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Life cycle assessment of a carbon capture utilization and storage supply chain in Italy and Germany: comparison between carbon dioxide storage and utilization systems

Grazia Leonzio*[1], I. David L. Bogle[2], Pier Ugo Foscolo[1]

[1]Department of Industrial and Information Engineering and Economics, University of L'Aquila, L'Aquila, 18 via G. Gronchi, 67100, Italy;

[2]Department of Chemical Engineering, Centre for Process Systems Engineering, University College London, London, Torrington Place WC1E 6BT, UK

*Corresponding author: grazia.leonzio@graduate.univaq.it

Abstract

The main purpose of this work is to verify that the CCUS supply chains at large scale that were developed in previous studies for Italy and Germany effectively reduce carbon emissions. The methodology of life cycle analysis was applied.

Results showed that the annual global warming potential (GWP) for these supply chains in Italy and Germany are respectively 9.62×10^{10} $kgCO_{2\text{-eq}}$ and 1.94×10^{11} $kgCO_{2\text{-eq}}$ which would help enable these countries to achieve the carbon dioxide reduction target fixed by European environmental policies. Overall emissions in Italy and Germany are 249 Mtonne/year and 640 Mtonne/year, respectively.

Sensitivity analysis results show that, for the supply chain in Germany, the GWP increases when, for a fixed amount of emissions captured, more carbon dioxide is sent to utilization: storage is then important to achieve the environmental target. Other impact categories decrease, increase or remain constant. On the other hand, for the supply chain in Italy, results showed that a lower environmental impact can be obtained by increasing the carbon utilization rate for methane production via a power to gas system. If this is implemented then this utilization system would a better solution from an environmentally point of view than the storage option with other utilization processes.

Keywords: CCUS supply chain, life cycle assessment analysis, carbon dioxide emissions, sensitivity analysis.

Abbreviations

ADP, abiotic depletion potential

AIMMS, advanced interactive multidimensional modeling system

AP, acidification potential

CED, cumulative energy demand

CCU, carbon capture and utilization

CCUS, carbon capture utilization and storage

CCS, carbon capture and storage

EP, eutrophication potential

FAETP, fresh water aquatic ecotoxity potential

GWI, global warming impact

GWP, global warming potential, equivalent to GWI

HTP, human toxicity potential

IGCC, integrated gasification combined cycle

LCA, life cycle assessment

LCI, life cycle inventory

LCIA, life cycle impact assessment

MAETP, marine aquatic ecotoxicity potential

MEA, monoethanolamine

MDEA, methyl diethanolamine

NGCC, natural gas combined cycle

ODP, ozone depletion potential

PC, pulverized coal

POCP, photochemical ozone creation potential

TETP, terrestrial ecotoxicity potential

1. Introduction

Global carbon dioxide concentration in the atmosphere has rapidly increased over the past decades at the rate of about 2 ppm/year and exceeded 400 ppm in 2016 (Kahn, 2016). In order to limit the rise of the global average temperature to 2 °C by 2050, the 2030 Climate and Energy Policy Framework proposed a reduction in carbon dioxide emissions of at least 40%, compared to the 1990 level, by 2030 (General Secretariat of the Council, 2014) and by 80-95% by 2050 (Pacala and Socolow, 2004). To achieve these aims, carbon supply chains have an important role and are among the research priorities of the Strategic Energy Technologies (SET) Plan of the European Union (European Commission, 2015 a,b).

Carbon capture and storage (CCS) supply chains, carbon capture utilization (CCU) supply chains and carbon capture utilization and storage (CCUS) supply chains have been proposed, as widely reported in the special issue of Zhang et al. (2020). In these technologies, carbon dioxide is captured from flue gases and is transported to a geological or ocean storage site and/or to a utilization site for its valorization with the production of valuable compounds (Mac Dowell et al., 2017). CCUS systems have the advantage of being a vital and potentially effective technology able to decrease emissions (Zhang et al., 2020).

Carbon supply chains require a significant amount of energy for their operation (especially for carbon dioxide capture and conversion processes) and this causes an additional environmental penalty. It was estimated that the increase in fuel consumption per kWh for plants that capture 90% of carbon dioxide by using the best current technologies ranges from 24 to 40% for new supercritical pulverized coal plants, from 11 to 22% for natural gas combined cycle plants, and from 14 to 25% for coal-fired integrated gasification combined cycle systems, compared to similar plants without capture and storage systems (IPCC, 2005). Therefore, it is necessary to know if a specific carbon supply chain is favorable from an overall environmental point of view. For this purpose, a life cycle assessment (LCA) analysis should be developed. LCA develops a series of metrics that consider all inputs and outputs in a process, analyzed over their entire life cycle (von der Assen et al., 2014a).

LCA studies have been conducted for CCS, CCU and CCUS supply chains in the literature. A number of LCA studies about CCS systems are reported in Table 1. An environmental benefit is not always ensured for this kind of supply chain. Kim et al. (2019) recommends that only 64% of carbon dioxide emitted by a power plant be sequestered and stored, while in Petrescu et al. (2017) a reduction for all environmental impact metrics is not obtained using a CCS supply chain. However, there are cases where the use of this framework does allow a reduction of the Global Warming Potential (GWP) and does result in a simultaneous increase of other impact metrics (Corsten et al., 2013; Singh et al., 2011a,b; Cuellar-Franca and Azapagic, 2015b). Other studies were focused on the evaluation of GWP and a significant reduction was predicted by Volkart et al. (2013), Ni et al. (2011), Koorneef et al. (2008) and Koore et al. (2010). In these studies carbon dioxide is mainly captured from a power plant. Only a few studies have reported benefits of a CCS framework for all investigated impact metrics (Pehnt and Henkel, 2009).

Table 1 Literature studies about LCA of CCS supply chains

Work	CO_2 source	Capture technology	Results - Effects of using a CCS supply chain
Cuellar-Franca and Azapagic (2015b)	Power plant	Post and pre combustion and oxy-fuel technology	Reduction of GWP by 63-82% per unit of generated electricity but with an increase of acidification and human toxicity
Volkart et al. (2013)	Fossil power plant and cement plant		Reduction of GWP (68-92% in fossil power plant and 39-72% in cement plant)
Pehnt and Henkel (2009)	Lignite power plant		Reduction of all impact categories in the pre-combusion technology
Singh et al. (2011a)	Natural gas combined cycle (NGCC)		Reduction of GWP by 64% with an increase of 43% in acidification, 35% in eutrophication and 120-170% in various toxicity impacts
Nie et al. (2011)	Power plant	Post-combustion and oxy-fuel technology	Reduction of GWP by 78.8% (post-combustion) 80% (oxy-fuel technology)
Koorneef et al. (2008)	Coal power plant		Reduction of GHGs up to 243 g/kWh
Koore et al. (2010)	Power plant		Reduction of GWP by 80%
Singh et al. (2011b)	Power plant		Reduction of GHGs by 64-78% but with an increase of toxicity
Corsten et al. (2013)	Fuel technology		Reduction of GWP but with an increase of eutrophication and acidification
Petrescu et al. (2017)	Coal power plant	MDEA, aqueous ammonia and calcium looping	No reduction for all impact categories
Kim et al. (2019)	Power plant	MEA absorption	Sequestration of only 64% of the emitted CO_2

LCA studies for CCU supply chains are shown in Table 2. Also for this kind of framework, an environmental benefit is not always ensured. In Passell et al. (2013), carbon dioxide is used to produce diesel from microalgae, but a GWP higher than the conventional petroleum based route is predicted. In Han and Lee (2013), carbon dioxide is utilized to produce polymers and bio-butanol, and a significant reduction of the environmental burden is ensured only by decreasing the gas-MEA capture. However, compared to the conventional production route, environmental advantages of using a CCU supply chain are reported in some studies where carbon dioxide is captured to produce dimethylcarbonate (Aresta and Galatola, 1999), polyols (Assen and Bardow, 2014a) and for mineral carbonation (Khoo et al., 2021).

Table 2 Literature studies about LCA of CCU supply chains

Work	CO_2 source	Capture technology	Utilization route	Results – Effects of using a CCU supply chain
Passell et al. (2013)	Electricity generation plant		Diesel from microalgae	Higher value of GWP (2.9 $kgCO_{2eq}$/1 MJ of combusted fuel) compared to the petroleum diesel (0.12 $kgCO_{2eq}$/1 MJ of combusted fuel)
Aresta and Galatola (1999)	Ammonia production plant	MEA absorption	Dimethyl carbonate	Reduction of GWP by 4.3 times compared to the conventional route from phosgene (31 vs 132 $kgCO_{2eq}$/kg dimethylcarbonate)
Khoo et al. (2021)	Flue gas from a waste-to-energy plant		Mineralization	Reduction of GWP by 115.78 $kgCO_{2eq}$ per tonne of CO_2 input
Han and Lee (2013)	Gas fired and coal fired power plants	MEA absorption	Polymers and bio-butanol	A significant reduction in the environmental impact is obtained by reducing the gas-MEA capture facilities
Assen and Bardow (2014b)	Lignite power plant		Polyols for polyurethane	Reduction of GHG by 11-19% and saving of fossil resource by 13-16% compared to the conventional route

LCA studies on CCUS supply chains are reported in Table 3. The utilization route is mainly arising from carbon dioxide oil recovery technology (CO_2-EOR). Studies show a GHG emission reduction of up to 80% (Hertwich et al., 2008). The attractiveness of the CCUS scheme was reported by Cooney et al. (2015). They showed that when the crude recovery ratio is increased emissions are reduced. Hussain et al. (2013) proposed different solutions for carbon dioxide source and capture and that this kind of CCUS scheme could result in negative emissions (Hornafius and Hornafius, 2015). When compared to conventional oil production, a CCUS framework based on CO_2-EOR can ensure up to 71% of emission reduction (Thorne et al., 2020; Azzolina et al., 2017; Abotalib et al., 2016).

In addition to the oil recovery process, other utilization options were considered for the environmental analysis of CCUS supply chains. In Yue and You (2015), carbon dioxide was stored and used to produce algae for biofuel production obtaining a reduction in emissions of up to 80%. Fernandez-Dacosta et al. (2018) studied the production of dimethyl ether and polyol obtaining lower values of GWP and fossil resource depletion.

Table 3 Literature studies about LCA of CCUS supply chains

Work	**CO_2 source**	**Capture technology**	**Utilization route**	**Results – Effect of using a CCUS supply chain**
Hertwich et al. (2008)	Power plant	MEA absorption	Oil recovery	Reduction of GHGs by 80%
Cooney et al. (2015)	Natural dome and power plant		Oil recovery	Reduction of emissions only for natural CO_2 when the crude recovery ratio is increased
Hussain et al. (2013)	Coal integrated gasification combined cycle (IGCC), switchgrass IGCC, livestock manure biogas natural gas combined cycle (NGCC), natural gas NGCC		Oil recovery	Coal and biomass IGCC CO_2-EOR, as well as natural gas and biogas NGCC CO_2-EOR, may be attractive alternatives for reducing GHG emissions
Hornafius and Hornafius (2015)	Corn ethanol fermentation		Oil recovery	Negative emissions are obtained
Azzolina et al. (2017)	Coal power plant		Oil recovery	Reduction of emissions compared to the conventional method of extraction
Jiang et al. (2017)	IGCC and pulverized coal (PC) power plant		Oil recovery	CO_2 emissions are 114.69-121.50 Mtonne $CO_{2\text{-}eq}$ (for IGCC), 222.95-236.19 Mt $CO_{2\text{-}eq}$ (for PC)
Abotalib et al. (2016)	Ethanol plant, coal-fired and natural gas fired power plant		Oil recovery	Reduction of carbon intensity compared to conventional crude recovery (up to -1.6 tonneCO_{2eq}/bbl for CO_2 from ethanol plant)
Thorne et al. (2020)	Power plant	Oxy-fuel technology	Oil recovery	Reduction by 71% of emissions compared to the conventional production of oil
Liu et al. (2020)			Oil recovery	Emissions of 2532.63 kg of CO_2, 74.18 kg of SO_2 and 37.38 kg of NOx per metric tonne of crude oil
Yue and You (2015)	Power plant		Algae for biofuel production	Reduction by 80% of CO_2 emissions when 187 Mgal of renewable diesel are produced
Fernandez-Dacosta et al. (2018)	Steam methane reforming unit		Dimethyl ether and polyol	Reduction of GWP and fossil depletion compared to the conventional production way (0.239 kgCO_{2eq}/FU and 0.131 kg_{oileq}/FU vs 0.294 kgCO_{2eq}/FU and 0.14 kg_{oileq}/FU)

This literature survey shows the importance of conducting an environmental analysis of each carbon supply chain to verify the effective reduction of emission notwithstanding any additional energy consumption. It is important to contrast the LCA with the economic analysis.

Other carbon-dioxide-based products have not been taken into consideration for the LCA of carbon supply chains and a comparison between carbon dioxide storage and new utilization options has not been analyzed so far in the literature for CCUS. Previous studies did not consider the application of LCA to CCUS supply chains at large scale (e.g. taking into account a supply chain developed for an entire Nation).

This study will contribute to fill these gaps. In particular, in previous work we analysed the CCUS supply chain for Germany (Leonzio et al., 2019) producing different carbon-dioxide-based products such as methanol, urea, concrete, wheat, polyurethane, calcium carbonate, lignin, and for Italy (Leonzio and Zondervan, 2020) producing methane. These were all at national scale considering the whole national territory. The aim of this research is to verify that each one of the systems optimized before is effectively able to reduce carbon dioxide emissions overall according to their respective national environmental targets. A considerable amount of energy is required for their operation: for the CCUS supply chain of Germany the energy consumption was of 55.37 GJ/ton CO_2 captured (Leonzio et al., 2019), while for the CCUS supply chain of Italy the energy consumption was of 28.8 GJ/ton CO_2 captured (Leonzio and Zondervan, 2020).

A sensitivity analysis has been developed for both supply chains to evaluate the influence of aspects of storage and utilization on the environmental results. A variable amount of the captured carbon dioxide can be sent to the utilization section to produce different compounds instead of being stored. This analysis can help the choice between carbon dioxide utilization or storage in order to create a more environmentally beneficial system.

The paper is divided into two parts: in the first part, the LCA of the CCUS supply chains in Italy and Germany is developed, while in the second part the environmental impact is evaluated through sensitivity analysis by increasing the utilization rate of carbon dioxide.

2. Materials and methods

LCA is a quantitative methodology used to evaluate the environmental impact of systems according to the standards ISO 14044 and ISO 14040 (ISO 14040, 2009; ISO 14044, 2006). Four important phases characterize this analysis, as shown in Figure 1: goal and scope, life cycle inventory (LCI) phase, life cycle impact assessment (LCIA) phase and interpretation phase (von der Assen et al., 2014a; ISO 14040, 2009; ISO 14044, 2006). To develop the LCA, GaBi software with the Ecoinvent database has been used (Education license, version 6) (Thinkstep, 2019).

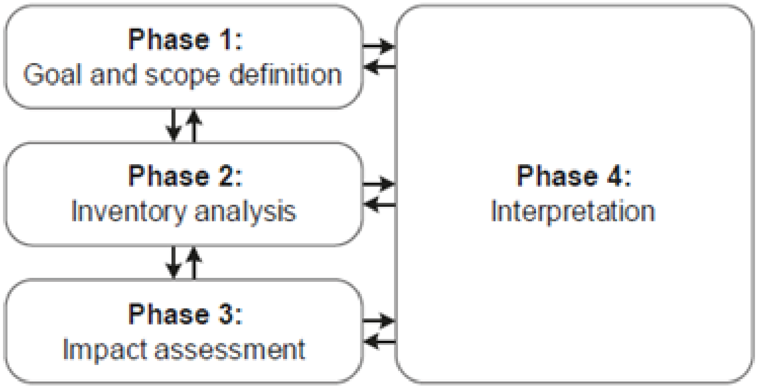

Figure 1 Stages of a life cycle assessment analysis according to the ISO standards (ISO 14040, 2009; ISO 14044, 2006)

2.1 Goal and scope

The goal of this study was to evaluate the environmental performances of the CCUS supply chains at national scale producing various products in Germany and methane in Italy (Leonzio et al., 2019; Leonzio and Zondervan, 2020). It was necessary to demonstrate that the suggested CCUS supply chains achieve the target set by European environmental policies especially for carbon dioxide emissions as defined in Gracceva et al. (2017) for Italy and in Ochoa Bique et al. (2018) for Germany. A sensitivity analysis was carried out to verify the influence of utilization and storage sections on the environmental impact. For a more complete analysis, different impact categories were considered: acidification potential (AP), eutrophication potential (EP), ozone depletion potential (ODP), abiotic depletion potential (ADP) fossil and elements, fresh water aquatic ecotoxicity potential (FAETP), human toxicity potential (HTP), marine aquatic ecotoxicity potential (MAETP), photochemical ozone creation potential (POCP), terrestrial ecotoxicity potential (TETP). Conditions ensuring higher sustainability have been identified.

2.1.1 Functional unit

The CCUS supply chains considered here have multi functionalities (von der Assen et al., 2014a). In order to define the functional unit, a system expansion methodology was applied. This allows the joint evaluation of all functions (where a function in LCA is expressing the recycling of carbon dioxide in different valuable products and/or the co-production of multiple valuable products): the functional unit was expanded to contain all functions (e.g. a sum of the single functions is considered according to the principle of the LCA for multi-functional problems) (Fernandez-Dacosta et al., 2018).

With these considerations, for the CCUS supply chain in Germany (Leonzio et al.,2019) the functional unit is the following, as defined in Table 4.

Table 4 Definition of the functional unit for the CCUS supply chain of Germany

Cement (Mtonne)	76.7
Electricity (kWh)	1.22×10^{11}
Iron and steel (Mtonne)	13.6
Concrete curing (Mtonne)	4.79
Wheat (Mtonne)	19.4
Lignin (Mtonne)	0.378
Polyurethane (Mtonne)	11
Calcium carbonate (Mtonne)	120
Urea (Mtonne)	1.34
Methanol (Mtonne)	0.846
Concrete by red mud (Mtonne)	19.2
Stored CO_2 (Mtonne)	20.3

For the CCUS supply chain in Italy (Leonzio and Zondervan, 2020) the functional unit is given in Table 5.

Table 5 Definition of the functional unit for the CCUS supply chain of Italy

Iron and steel (Mtonne)	66.92
Electricity (kWh)	2.25×10^{10}
Methane (Mtonne)	16.1
Stored CO_2 (Mtonne)	32.8

As Table 4 and 5 show, a definition of functional unit in absolute terms was considered for this work.

2.1.2 System boundaries

Cradle-to-gate analyses were performed for the supply chains (the use and disposal phases of carbon-dioxide-based products were not considered in the LCA). Carbon dioxide was considered as a feedstock (economic flow) and not only as an emission (von der Assen et al., 2014a). System boundaries describing which processes of the supply chain are included in the assessment for the CCUS framework in Germany are shown in Figure 2. The CCUS supply chain of Germany in this environmental analysis is that described in the work of Leonzio et al. (2019). The location of carbon dioxide source and utilization sites was fixed.
According to the economic optimization the selected carbon dioxide sources for the whole national territory were: Dresden, Wiesbaden, Berlin, Munich, Potsdam, Magdeburg, Saarbrucken. These are representative of different kind of industries producing flue gas.

Upstream of the system boundaries three different inputs are present: power plants (Wiesbaden, Berlin, Potsdam), cements plants (Dresden, Saarbrucken) and steel and iron plants (Munich, Magdeburg). Downstream of the storage and utilization processes, where the captured carbon dioxide is used or stored, nine output streams are present, namely different products of carbon dioxide utilization routes (methanol, urea, concrete curing, concrete by red mud, wheat, polyurethane, calcium carbonate, lignin) and the stored carbon dioxide.

The storage site is located in Altmark, the concrete curing sites in Ennigerloh and Hannover, the wheat cultivation sites in Munich and Hannover, the lignin utilization sites in Cologne and Münchsmünster, the polyurethane production sites in Schwarzheide and Leverkusen, the calcium carbonate production sites in Salzgitter and Bremen, the urea production sites in Kassel and Hagen, the methanol production sites in Leuna and Wesseling, and the concrete production sites by red mud in Rackwitz and Hamburg (the amount of each carbon dioxide based product and the amount of stored carbon dioxide is that provided in the functional unit).

The utilization routes were chosen according to the potential for different utilization options of carbon dioxide in Germany, as suggested in the literature (Patricio et al., 2017). Carbon dioxide capture and compression and carbon dioxide transportation were considered to be within the up- and downstream processes.

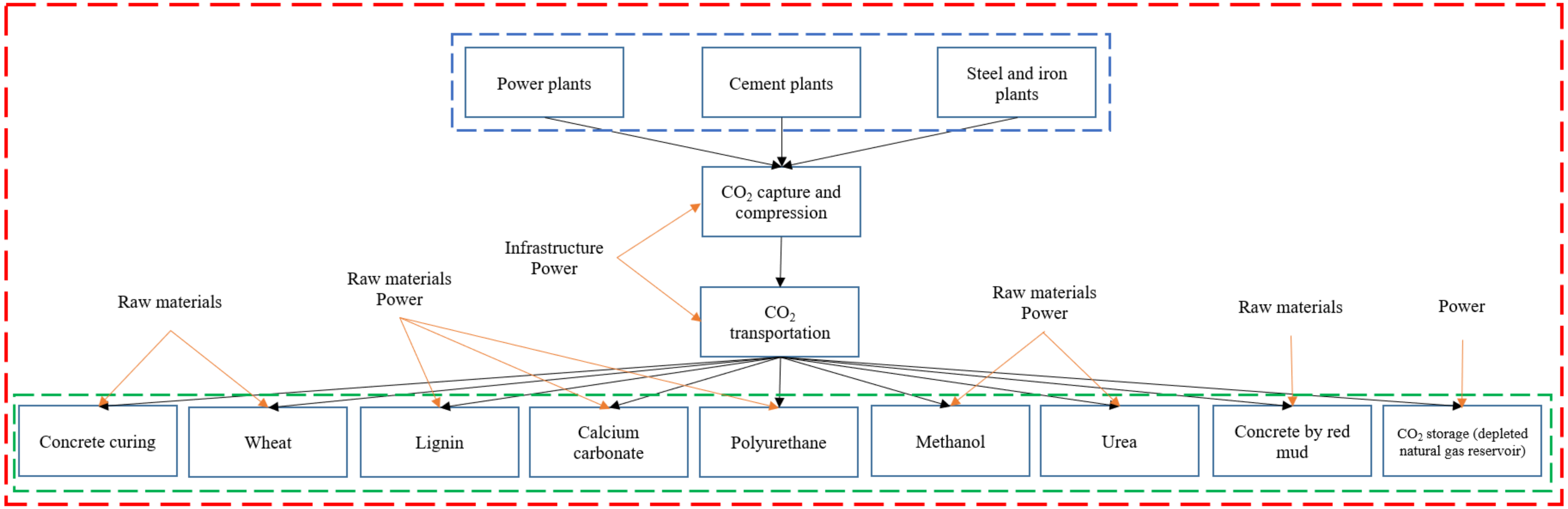

Figure 2 System boundaries for the CCUS supply chain in Germany - red line: system boundaries; green line: downstream processes (production of CO_2-based compounds and CO_2 storage); blue line: upstream processes

Figure 3 shows the system boundaries for the CCUS supply chain in Italy. The CCUS supply chain of Italy is that described in the work of Leonzio and Zondervan (2020) where carbon dioxide sources and the utilization site are fixed as a result of the economic optimization. Throughout the whole national territory, the selected carbon dioxide sources for the optimal supply chain are the following: Puglia, Lombardy, Emilia Romagna and Piedmont, with different types of industry producing flue gas.
Inside the system boundaries, flue gas sources are present upstream: a power plant (Emilia Romagna) and iron and steel plants (Puglia, Lombardy, Piedmont). Downstream of the utilization process and storage section, methane and stored carbon dioxide are considered. The utilization site is located in Verbania while the storage site is offshore in the Adriatic sea saline aquifer.

The potential for the application of power to gas plants in Italy was reported by Colbertaldo et al. (2018) and Guandalini et al. (2017). Moreover, the production of methane is specifically selected for the Italian regions because the potential of methanol production is very low there (Patricio et al., 2017). On the other hand, Italy would have the opportunity to satisfy its national methane demand by hydrogenation of CO_2. In addition, an existing network for CH_4 distribution is present in Italy and a power-to-gas system is achievable on a large scale with a high technical readiness level. In this case carbon dioxide capture, compression and transportation are included between the upstream and downstream processes.

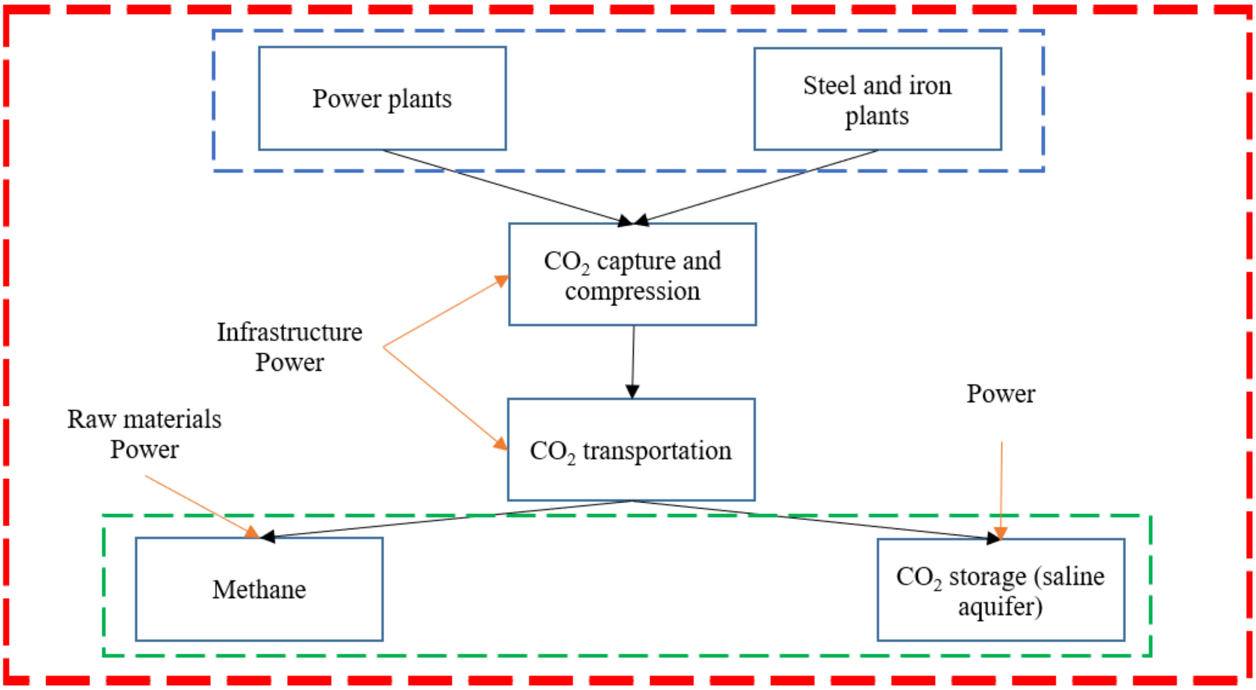

Figure 3 System boundaries for the CCUS supply chain in Italy - red line: system boundaries; green line: downstream processes (production of methane and CO_2 storage); blue line: upstream processes

For both supply chains the utilization options chosen were the most economically appealing carbon-dioxide–based products for the respective countries. Only methane is taken into account in the framework for Italy, while methanol, urea, concrete curing, concrete by red mud, wheat, polyurethane, calcium carbonate, lignin are proposed for the German supply chain.

In both supply chains system boundaries included the construction of plants related to carbon dioxide sources, the extraction or production of raw materials and their transportation calculated using the database in the GaBi software (Thinkstep, 2019). The production and transport of raw materials for the infrastructure needed for carbon dioxide pipelines were also considered (Koornneef et al., 2008). Infrastructure needs were considered only for carbon dioxide capture with MEA absorption due to the scarce availability of data for different capture technologies (Giordano et al., 2018). For the utilization processes infrastructure data was not available. However, the production of raw materials was considered inside the system boundary. Natural infrastructures to store CO_2 are available in both cases examined here (a saline aquifer and a depleted natural gas reservoir). Only power needed for the storage process is taken into account (Wildbolz, 2007).

2.2 Life cycle inventory phase

In the LCI phase, input and output data for all processes of the CCUS supply chains were provided. The results for the large-scale supply chains obtained by the optimization were used (Leonzio et al., 2019; Leonzio and Zondervan, 2020). Based on this process data, an inventory list for the complete life cycle was calculated. All elementary flows entering the process (technosphere) from nature (environment) in the form of resources and those leaving the process in the form of resources, deposited goods, emissions to air, fresh water, sea water, agricultural and industrial soil were taken into account, as shown in Figure 4.

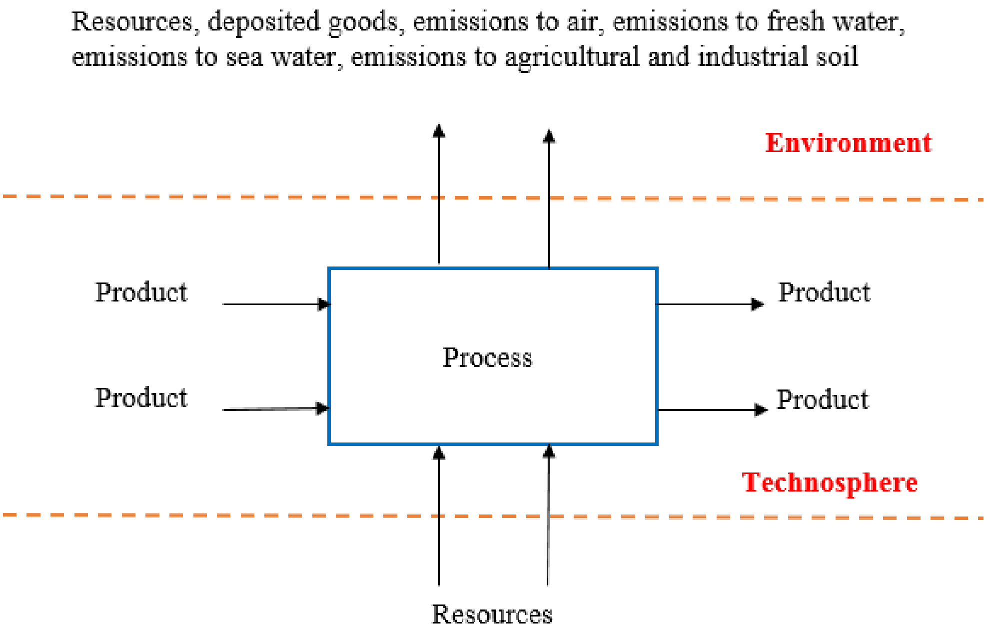

Figure 4 Elementary flows entering and leaving the process

For the CCUS supply chain of Germany (Leonzio et al., 2019), an inventory analysis was performed for production from carbon dioxide of methanol, calcium carbonate, polyurethane, urea, wheat, concrete curing, concrete by red mud and lignin treatment with carbon dioxide. Tables 6 and 7 show respectively the inventory analysis for methanol production by carbon dioxide hydrogenation and for hydrogen production by water electrolysis, using the work of Biernacki et al. (2018), Michailos et al. (2018), Kajaste et al. (2018), and Matzen and Demirel (2016). The recycle of unconverted gases to the methanol reactor after separation of water and methanol was also considered with an efficiency of 80%. In summary, 1.7 tonne of carbon dioxide are consumed per ton of produced methanol (Patricio et al., 2017). Catalyst (aluminum, copper and zinc oxide) is required (as an input) but it is not consumed.

Table 6 Inventory analysis for methanol synthesis via carbon dioxide hydrogenation in the CCUS supply chain of Germany (Biernacki et al., 2018; Michailos et al., 2018; Kajaste et al., 2018; Matzen and Demirel, 2016; Patricio et al., 2017)

Input of the process		
H_2	0.23	tonne
CO_2	1.70	tonne
Aluminum oxide	0.01	tonne
Copper oxide	0.08	tonne
Zinc oxide	0.04	tonne
Water	5.45	tonne
Energy	0.13	MWh
Output of the process		
Wastewater	0.68	m^3
CO_2	0.34	tonne
Methanol	1	tonne

Table 7 Inventory analysis for the electrolyzer in the methanol production process in the CCUS supply chain of Germany (Biernacki et al., 2018; Michailos et al., 2018; Kajaste et al., 2018; Matzen and Demirel, 2016; Patricio et al., 2017)

Input of the process		
Water	2.22	tonne
Traditional energy	141.96	kWh
Renewable energy	6.11	kWh
Output of the process		
Oxygen	1.97	tonne
H_2	0.23	tonne
Waste water	0.12	m^3
VOC	5.23	g
CO	47.67	g
NOx	41.31	g
PM_{10}	15.16	g
$PM_{2.5}$	7.50	g
SOx	276.67	g
CH_4	47.06	g
CO_2	28.22	kg
SF6	0.92	mg
C_2F_6	0.10	g
Black carbon	0.25	g
POC	0.48	g

Table 8 shows the inventory analysis for calcium carbonate production from carbon dioxide based on Mattila et al. (2014) and Zappa (2014) and considering that the ratio between each tonne of used carbon dioxide and a tonne of steel slag is 0.42. An inventory analysis for lignin treatment with carbon dioxide is presented in Table 9 based on Bernier and Lavigne (2013) and considering that 0.22 tonne of carbon dioxide per ton of lignin are utilized (Patricio et al., 2017).

Table 8 Inventory analysis for the calcium carbonate production process from carbon dioxide in the CCUS supply chain of Germany (Mattila et al., 2014; Zappa, 2014)

Input of the process		
Steel slag	2.60	tonne
CO_2	1.1	onne
NH_4Cl solvent	0.03	tonne
Electricity	107.40	kWh
Water	2.10	tonne
Steam	37500	MJ
Output of the process		
Calcium carbonate	1.00	tonne
Waste water. steel slag	2.01	tonne

Table 9 Inventory analysis for the lignin treatment process with carbon dioxide in the CCUS supply chain of Germany (Bernier and Lavigne; 2013; Patricio et al., 2017)

Input of the process		
Natural gas	0.46	tonne
CO_2	0.22	tonne
H_2SO_4	0.17	tonne
NaOH	0.08	tonne
$CaCO_3$	0.17	tonne
Water	3.56	tonne
Electricity	7.33	kWh
Output of the process		
SO_2	6.75×10^{-9}	tonne
NO_x	3.38×10^{-7}	tonne
CO	3.82×10^{-7}	tonne
Lignin	1	tonne

Polyurethane is produced from polyols and isocyanate. The first reactant is obtained from carbon dioxide, while the latter one is obtained from carbon monoxide produced by methane steam reforming. All inputs and outputs for these processes are shown in Table 10 based on von der Assen et al. (2015). For polyurethane production from carbon dioxide, the ratio between used carbon dioxide and polyurethane is 0.3 tonne/tonne (Patricio et al., 2017). More information about the polyurethane production route that is taken into account here is available in the Supplementary Material.

Table 10 Inventory analysis for the polyurethane production from carbon dioxide in the CCUS supply chain of Germany (Von der Assen et al., 2015; Patricio et al., 2017)

Polyurethane production (flexible foam)		
Input of the process		
Polyols	0.713	kg
Electricity	1.5	MJ
TDI	0.285	kg
Output of the process		
Flexible foam	1	kg
GW	0.051	kg$CO_{2\text{-eq}}$
Isocianate production		
Input of the process		
Toluene	0.15	kg
Electricity	3.77	MJ
CO	0.09	kg
Nitirc acid	0.21	kg
Output of the process		
TDI	0.285	kg
Steam reforming		
Input of the process		
CH_4	0.066	kg
Electricity	0.353	MJ
Heat	0.747	MJ
Output of the process		
H_2	0.020	kg
CO	0.092	kg
Polyols production		
Input of the process		
Starter	0.019	kg
Propylen oxide	0.395	kg
CO_2	0.299	kg
Output of process		
Polyols	0.713	kg

Table 11 Inventory analysis for urea production from carbon dioxide in the CCUS supply chain of Germany (Antonetti et al., 2017; Patricio et al., 2017)

Input of the process			
NH_3		0.57	tonne
CO_2		0.74	tonne
Energy		0.05	MWh
Output of the process			
NH_3 emissions		0.004	tonne
Wastewater		0.48	tonne
	NH_3	0.03	tonne
	CO_2	0.02	tonne
	Urea	0.005	tonne
	water	0.43	tonne
Urea		1	tonne

An inventory analysis for urea production from carbon dioxide is shown in Table 11 based on Antonetti et al. (2017). Here 0.74 tonne of carbon dioxide are used to produce 1 tonne of urea (Patricio et al., 2017).

Inventory analyses for concrete produced by red mud and concrete curing are shown respectively in Tables 12 and 13 using data reported by Nikbin et al. (2018) and Gursel and Horvath (2012). The GWP and cumulative energy demand (CED) are respectively 330.74 kgCO_{2-eq} and 2848.5 MJ for concrete production by red mud (1 tonne) (Nikbin et al., 2018). The GWP and CED are respectively 292 kgCO_{2-eq} and 1374.68 MJ for concrete curing (1 tonne) (Nikbin et al., 2018; www.carboncure.com). In concrete curing, 0.03 tonne of carbon dioxide are required for 1 tonne of concrete (Patricio et al., 2017), while in concrete by red mud production the ratio between one tonne of carbon dioxide and one tonne of red mud is 0.17.

Table 12 Inventory analysis for concrete production by red mud and carbon dioxide in the CCUS supply chain of Germany (Nikbin et al., 2018; Gursel and Horvath, 2012; Patricio et al., 2017)

Input of the process		
Cement	0.222	tonne
Red mud	0.074	tonne
Coarse Agg	0.129	tonne
Fine Agg.	0.106	tonne
Leca	0.197	tonne
Limestone	0.118	tonne
Water	0.150	tonne
Superplasticizer	0.004	tonne
CO_2	0.013	tonne
Output of the process		
Concrete	1	tonne
CO	0.04619	tonne
Lead	0.000012	tonne
NO_X	0.00075	tonne
PM_{10}	0.00002	tonne
SO_2	0.00076	tonne
VOC	0.00057	tonne

Table 13 Inventory analysis for concrete curing in the CCUS supply chain of Germany (Nikbin et al., 2018; Gursel and Horvath, 2012; Patricio et al., 2017)

Input of the process		
Cement	0.285	tonne
Water	0.145	tonne
Fine aggregates	0.118	tonne
Coarse aggregates	0.145	tonne
Leca	0.190	tonne
Limestone	0.114	tonne
Superplasticize	0.003	tonne
CO_2	0.030	tonne
Output of the process		
CO	0.056	tonne
Lead	0.00001	tonne
Nox	0.001	tonne
PM_{10}	0.00003	tonne
SO_2	0.001	tonne
VOC	0.001	tonne
Concrete	1.000	tonne

CO_2 can also be utilized at large scale in agricultural processes. The environmental burden and inventory analysis for wheat growing enhanced by carbon dioxide is shown in Table 14 based on Biswas et al. (2010, 2008). According to tests of free air concentration enrichment (FACE) of carbon dioxide performed around the world, this utilization route is not as effective as preliminary laboratory experiments showed. CO_2 concentration cannot be increased much above its value in the atmosphere (Erda et al. 2005) because the absorption rate of carbon by photosynthesis increases at the expenses of Nitrogen (proteins) and additional nutrients changing the quality of biomass grown. For this reason, the contribution of CO_2, in addition to that already available in the atmosphere, can be only marginal and was evaluated as 500 mg per kg of wheat produced (Leonzio et al., 2019).

Table 14 Inventory analysis for wheat production in the CCUS supply chain of Germany (Biswas et al., 2010; 2008; Leonzio et al., 2019)

Input of the process		
CO_2	0.5	kg
Output of the process		
Wheat	1	tonne
GW	275	$kgCO_{2\text{-eq}}$

In the Italian CCUS supply chain (Leonzio and Zondervan, 2020) methane is produced from carbon dioxide with a power to gas process (by means of hydrogen produced by water electrolysis exploiting renewable energy sources). Table 15 shows the inventory analysis for the methanation process (using the Sabatier reaction) assuming complete CO_2 conversion to methane (Reiter et al., 2015; Sternberg and Bardow, 2016).

Table 15 Inventory analysis for the methanation process (Sabatier reaction) in the CCUS supply chain of Italy (Reiter et al., 2015; Sternberg and Bardow, 2016)

Input of the process		
CO_2	2.75	kg
H_2	0.5	kg
Electricity	0.33	kWh
Output of the process		
CH_4	1	kg
Waste heat	8.26	MJ
H_2O	2.29	kg

In both CCUS supply chains considered previously (Leonzio et al., 2019; Leonzio and Zondervan, 2020) carbon dioxide was also stored: it was assumed that electrical energy is used to inject carbon dioxide and the required energy is $2.86 \cdot 10^{-2}$ $kWh/kgCO_2$ (Wildbolz, 2007). Regarding carbon dioxide transportation, infrastructures and energy, data were used from Koornneef et al. (2008) and Wildbolz (2007), respectively. It was assumed that carbon dioxide recompression is necessary for a distance exceeding 400 km and that the associated energy consumption is 0.011 kWh/(tonne km) (Wildbolz, 2007). No leakage emissions were considered due to their negligible value (Bouman et al., 2015).

For the inventory analysis of carbon dioxide capture with piperazine absorption, the energy requirement was estimated: von der Assen et al. (2015) suggested a value of 0.80 GJ/tonneCO_2, while 0.86 GJ/tonneCO_2 are necessary for the absorption of carbon dioxide with MEA. Infrastructure and emissions data for this last technology were proposed by Giordano et al. (2018).

The LCI results (using GaBi software) for the German CCUS supply chain (Leonzio et al., 2019) are classified as input (resources) and output (resources, deposited goods, emissions to air, fresh water, sea water, agricultural soil and industrial soil). In the output the greatest contribution to elementary flows is made by the emissions to fresh water (63.4%) followed by the emissions to air (35.8%). Other terms are negligible. These elementary flows can be expressed also in kg$CO_{2\text{-eq}}$. In the input 5.24×10^{11} kg$CO_{2\text{-eq}}$ are present. In the output, 7.08×10^{11} kg$CO_{2\text{-eq}}$ of emissions to air are produced. Steam production in the calcium carbonate process and carbon dioxide source in Munich provides the highest contribution to the emissions to air.

For each carbon dioxide source, carbon dioxide (as a feedstock sent to storage or utilization) is co-produced with the main product of the industry emitting flue gas. To allocate the environmental impact between the main product and the co-product, the price allocation method was adopted. The prices for carbon dioxide, electricity, cement and steel are respectively 80 €/tonne, 0.15 €/kWh, 80 €/tonne and 589 €/tonne (Focus Economics, 2019; Boyer and Ponssard, 2013; Europe, 2019; OECD, 2013).

The LCI results were also obtained using the GaBi software for the Italian CCUS supply chain (Leonzio and Zondervan, 2020). The greatest impact was due to the emissions to fresh water and to air that contributed respectively 75.4% and 20.9% to the total output flow. The deposited goods contributed 2.97% to the total flow, while other contributions were negligible. Inputs and outputs were expressed also as kg$CO_{2\text{-eq}}$: input resource was 2.83×10^{8} kg$CO_{2\text{-eq}}$ while emission to air in the output was 9.67×10^{10} kg$CO_{2\text{-eq}}$. The highest contribution was due to the carbon dioxide sources in Lombardy and Puglia (iron and steel plants).

Also in this case a price allocation criterion was applied to carbon dioxide sources. The price taken into account for CO_2, electricity and steel are respectively 80 €/tonne, 0.15 €/kWh and 589 €/tonne (Portdata, 2019; Focus Economics, 2019; OECD, 2013).

3. Results

The results from the LCA are discussed in this section for both CCUS supply chains. The magnitude of the environmental burden was evaluated through two different steps: classification and characterization. In these steps the LCI results were combined and organized into the impact categories (classification step) and then into the impact indicators at the midpoint level of the cause-effect chain that analyzes the environmental effect due to defined causes (characterization step) using the CML 2001 methodology (Guinee et al., 2002) implemented in GaBi (Thinkstep, 2019) (LCIA phase). In the last phase of this environmental analysis, the interpretation of previous results in terms of significant issues and sensitivity analysis was developed (interpretation phase).

3.1 Life cycle impact assessment phase for the German CCUS supply chain

The environmental impact category that was considered is the Global Warming Potential (GWP), sometimes also referred to as the GWI or Global Warming Impact (Heijungs, 2014), because the first aim of this analysis was to determine the ability to meet the targets set by the national environmental policies with respect to carbon dioxide emissions. Results for the other impact categories such as AP, EP, ODP, ADP fossil and elements, FAETP, HTP, MAETP, POCP, and TETP are shown in the Supplementary Materials (see Table S1).

Results showed that the value for GWP for the German supply chain was 1.94×10^{11} $kgCO_{2\text{-eq}}$: the CCUS supply chain is able to achieve the target set by the German environmental policy for 2020 as their Government's environmental regulations. The German Federal Ministry for the Environment (2017) stated that in Germany carbon dioxide emissions should be lower than 751 $MtonneCO_2$ by 2020. On the other hand, for the CCUS of Germany described in Leonzio et al. (2019), total carbon dioxide emissions were 640 $MtonneCO_2$ which also includes emissions from sources not included in the optimized chain.

The greatest carbon dioxide emissions in the CCUS supply chain come from the carbon dioxide source in Munich (steel and iron plant) with 7.85×10^{10} $kgCO_{2\text{-eq}}$, followed by steam production in the precipitated calcium carbonate process, with 1.95×10^{10} $kgCO_{2\text{-eq}}$, in Potsdam (power plant) with 1.65×10^{10} $kgCO_{2\text{-eq}}$, from the process for the production of propylene oxide in polyurethane production with 1.53×10^{10} $kgCO_{2\text{-eq}}$, and by the process to produce concrete by red mud with 6.42×10^{9} $kgCO_{2\text{-eq}}$.

3.2 Life cycle impact assessment phase for the Italian CCUS supply chain

For the Italian CCUS supply chain (Leonzio and Zondervan, 2020) the goal of the analysis was again to ensure that the supply chain reduces carbon dioxide emissions to a value lower than that established by the national environmental policy, in this case equal to 275 $MtonneCO_2$ (Gracceva et al., 2017). For this reason, only the GWP impact category is considered. Results showed that for this CCUS supply chain the value of GWP was 9.62×10^{10} $kgCO_{2\text{-eq}}$. When considering also the additional carbon dioxide sources not included in the optimized supply chain the total carbon dioxide emissions were 249 $MtonneCO_2$. A value lower than the target was obtained showing that the proposed approach is able to effectively reduce carbon dioxide emissions in Italy. Inside the supply chain, the processes with the greatest contribution to carbon dioxide emissions are the iron and steel plant in Puglia (6.32×10^{10} $kgCO_{2\text{-eq}}$) and the iron and steel plant in Lombardy (1.09×10^{10} $kgCO_{2\text{-eq}}$). For a complete analysis, other impact categories such as AP, EP, ODP, ADP fossil and elements, FAETP, MAETP, POCP, TEP are reported in the Supplementary Materials (see Table S2).

4. Discussion

4.1 Interpretation phase for the German CCUS supply chain

The interpretation phase involves a sensitivity analysis around the base case to explore the extent to which the results are significant and the way that changes may affect them. The results in section 3.1 show that the significant processes that contribute most to the overall result were: the precipitated calcium carbonate process, due to the high environmental impact of steam production, carbon dioxide sources (in particular Munich, Potsdam), propylene oxide formation in the polyurethane production section and concrete production by red mud.

A sensitivity analysis was carried out to evaluate the relative influence of the storage and utilization sections within the supply chain. For carbon dioxide captured from Magdeburg the amount that is sent to the storage section is used at different percentages for the production of different species (concrete by red mud or curing, wheat, lignin upgrading, urea, methanol, polyurethane and calcium carbonate). For three different case studies respectively 25%, 50% and 75% of the carbon dioxide originally stored in the base case was maintained in the storage section while the remaining carbon dioxide captured was sent to utilization in order to increase the production of the single product.

Results giving the negative or positive change are shown in Table 16 (the absolute values corresponding to the arrows of Table 16 are shown in Tables S3-S10 of the Supplementary Materials). For a complete picture of the environmental impact, all impact categories were considered (Supplementary Material). While the GWP is expected to be increased by increasing the utilization rate of carbon dioxide, other impacts could be increased, constant or decreased (Cuellar-Franca and Azapagic, 2015).

Table 16 Sensitivity analysis: trends resulting for different impact categories when increasing the carbon dioxide utilization rate in each utilization section of the supply chain for Germany; arrows indicate variation in direction and intensity for each impact category with reference to the base case (↑ / ↓ low variation (<5%), ⇔ constant value, ⇧ / ⇩ medium variation (<50%), ⬆ / ⬇ high variation (>50%))

	GWP	AP	EP	ODP	ADP elements	ADP fossil	FAETP	HTP	MAETP	POCP	TETP
Methanol	↑	⇔	⇔	⇔	↑	↓	↑	↓	⇔	⇔	↓
Concrete curing	⇧	⇧	↑	⇔	⇔	↓	↓	↑	⇔	⬆	↑
Urea	⇧	⇧	↑	↑	↑	⇧	↓	↑	⇔	↑	⇔
Wheat	⬆	⇔	⇔	⇔	⇔	↓	↓	↓	⇔	⇔	↓
Lignin treatment	↑	↑	⇔	↑	⇧	⇧	⇔	↑	↑	↑	⇔
Polyurethane	⇧	⇧	↑	⇧	⬆	⇧	↑	⇧	↑	↑	↑
Calcium carbonate	↑	↑	⇔	⇔	↑	↓	⇔	⇧	⇧	↑	⇧
Concrete by red mud	⇧	⇧	↑	⇔	⇔	↓	⇔	⇧	⇔	↑	⇧

The sensitivity analysis suggests that the storage site is important for the German CCUS supply chain in order to reduce the environmental impact in terms of GWP. In all cases the GWP was raised by increasing the amount of carbon dioxide sent to the utilization section while keeping constant the amount of captured carbon dioxide. Few impact categories were reduced and only for some carbon-dioxide-based products.

Comparing different case studies, a lower overall environmental impact was obtained when additional methanol was produced. The highest GWP value was obtained with wheat production. The higher environmental impact in terms of GWP at a higher utilization rate of carbon dioxide was due to a higher environmental impact for the utilization processes compared to that of the storage system. This result was in agreement with the work of Cuellar-Franca and Azapagic (2015) where a comparison between CCS and CCU was presented: on average the GWP for CCS is significantly lower than that for the CCU option. For example, for biodiesel production the GWP is four times higher than that for CCS, while the carbon mineralization and the EOR have a GWP that is 2.9 and 1.8 times higher than that for CCS, respectively. Even if the utilization solution produces a better economic return its overall environmental impact in terms of GWP compared to storage is higher.

This demonstrates that the storage section is important and should be designed at the optimal operating conditions. Also, storage is important because the demand for chemicals and other products does not have the capacity to sink enough carbon dioxide emissions to achieve the carbon reduction targets (Cuellar-Franca and Azapagic, 2015). It was estimated that the annual production of urea and methanol requires only 0.5% of the current 34.5 Gtonne/year of the anthropogenic global carbon dioxide emissions (ISPRA, 2013). The same conclusion about the importance of carbon storage was reported by Aldaco et al. (2019) comparing a CCU with a CCS system.

For the other impact categories considered here in the LCA and sensitivity analyses in addition to GWP, a comparison with the literature cannot be performed because a complete LCA, i.e. including the additional impact categories, was not yet developed in previous studies for CCUS supply chains. Cuellar-Franca and Azapagic (2015) suggested that a wider range of LCA impacts be considered in future rather than focusing only on the GWP and to examine the various utilization options of carbon dioxide as has been done for the supply chains here. However, as Table 16 shows, and as discussed above, some impact categories are increasing while others are decreasing or remain constant when raising the utilization rate of carbon dioxide to obtain different products.

Although LCA results for new utilization processes will undoubtedly evolve, LCA at this stage will help to provide suggestions for future studies aiming at higher energy efficiencies and other environmental advantages. With this in mind, an additional LCA study was carried out incorporating a higher efficiency of methanol synthesis in terms of global carbon dioxide conversion. This should reduce carbon dioxide emissions at the outlet of the chemical reactor (Leonzio and Foscolo, 2020; Leonzio et al., 2019). As defined before, the methanol reactor includes the recycle of unconverted gases after the separation of methanol and water to improve carbon dioxide conversion. Efficiencies higher than that corresponding to 80% recycle (used above in the base case) were considered here in order to study the effect of changing the carbon dioxide to methanol conversion rate on the various impact categories of the LCA for different utilization and storage rates of CO_2. Results of this sensitivity analysis are shown qualitatively in Table 17 (the exact values are reported in Tables S11 and S12).

Table 17 Results of sensitivity analysis when increasing carbon dioxide utilization rate at different rates of carbon dioxide conversion to methanol; arrows indicate variation of each impact category with reference to the base case (↔constant value;↑/↓low variation (< 5%) upwards/downwards).

CO_2 conversion	GWP	AP	EP	ODP steady state	ADP elements	ADP fossil	FAETP	HTP	MAETP	POCP	TETP
100%	↔	↔	↔	↔	↑	↓	↑	↓	↔	↔	↓
90%	↔	↔	↔	↔	↑	↓	↑	↓	↔	↔	↔

When increasing the utilization ratio of carbon dioxide, the GWP remains constant for a fixed amount of captured carbon dioxide and for a defined carbon dioxide conversion. On the other hand, for a different conversion rate the analysis resulting from increasing the utilization rate of carbon dioxide to methanol produces a different value of GWP with respect to the base case.

When global carbon dioxide conversion of methanol synthesis is fixed at 90% ADP elements and FAETP increased while ADP fossil and HTP decreased. When global carbon dioxide conversion is approaching 100% TETP also decreased, suggesting a lower environmental impact.

These results show that improving the efficiency of the methanol process allows a higher amount of carbon dioxide to be sent to the utilization section and the amount sent to the storage section to be reduced without increasing the value of GWP. However, it is difficult to operate a methanol process based on carbon dioxide hydrogenation with these high efficiencies. This result indicates that research towards increased methanol conversion rates would produce environmental and energetic advantages for CCUS.

4.2 Interpretation phase for the Italian CCUS supply chain

Previous results suggested that hotspots (processes with a higher environmental impact) were linked to CO_2 sources, especially in Puglia and Lombardy where iron and steel plants are present.

As for the German supply chain, a sensitivity analysis was undertaken keeping constant the amount of captured carbon dioxide while increasing the amount of emissions utilized for methane production and reducing those sent to storage. 25%, 50%, 75% of base case values for carbon dioxide sent to storage section were used in this analysis. The remainder was used for methane production. The amount of methane produced in the three sensitivity cases were 25 Mtonne/year, 22 Mtonne/year and 19 Mtonne/year. In the base case, 16.1 Mtonne/year of methane were produced. Results showed that when an increasing amount of carbon dioxide is sent to the utilization section the GWP was reduced. For the Italian supply chain utilization is preferred over storage because a lower GWP can be obtained, as shown in Figure 5, and for this reason the GWP associated with carbon dioxide storage by injection in the saline aquifer is higher than that of methane production. This may be explained by the noticeable utilization of hydrogen together with CO2 in the synthesis of methane (at a molar ratio of 4:1).

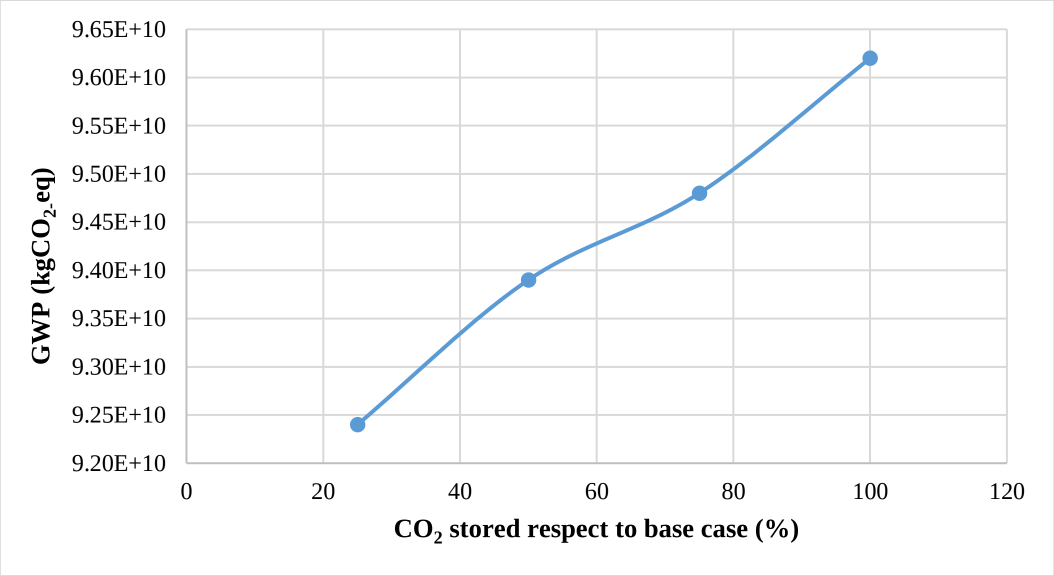

Figure 5 GWP as a function of different percentages of carbon dioxide sent to storage, with respect to the base case when additional methane is produced in the Italian CCUS supply chain

The trend for the other impact categories obtained when increasing the utilization rate of carbon dioxide (still keeping constant the amount captured) is shown in Table 18 (the exact values are reported in Table S13). Overall, a lower environmental impact was obtained because all impact categories were reduced except for EP which remained constant. In this case storage was an unfavorable option for reducing the environmental impact. These results suggest that power to gas technology is a cleaner and more environmentally friendly process than the storage option with other utilization systems which was the conclusion for the German case. No carbon dioxide emissions were present at the outlet of the chemical reactor. Carbon dioxide conversion and methane selectivity can reach values close to 100% especially under stoichiometric conditions (Stangeland et al., 2015).

Table 18 Results of the sensitivity analysis regarding different impact categories when higher fractions of CO_2 flow rate are utilized for methane production; arrows indicate variations with respect to the base case (↔ constant value, ↓/↑ low variation (<5%), ⬇/⬆ medium variation (<50%))

GWP	AP	EP	ODP steady state	ADP elements	ADP fossil	FAETP	MAETP	POCP	TETP
↓	↓	↔	⬇	↓	↓	↓	⬇	↓	⬇

4.3 Comparison and further discussion about the CCUS supply chains

The sensitivity analysis in section 4.1 showed that storage is preferred over utilization. For the Italian CCUS supply chain, utilization is preferred over storage to reduce the environmental impact (as discussed in section 4.2). These results suggest that the best carbon dioxide utilization system is the power to gas system. Sternberg and Bardow (2016) suggested that for power-to-gas the global warming impact is about 0.222 kg $CO_{2\text{-eq}}/FU_{SNG}$

while the fossil depletion impact is 0.072 kg Oil$_{-eq}$/FU$_{SNG}$ in 2020. Our results confirm that this technology is the most effective and mature process. It avoids an increase in the environmental impact for CO_2 utilization. While power to gas systems are expected to have an important role in the energy transition there are only few studies reporting LCA for this technology (Gotz et al., 2016; Meylana et al., 2017; Sternberg and Bardow, 2015).

5. Conclusions

In this study a Life Cycle Analysis was carried out for large scale CCUS supply chains developed in previous studies for Germany and Italy (Leonzio et al., 2019; Leonzio and Zondervan 2020). This study particularly incorporated the utilization of CO2 through its chemical conversion to a range of useful products.

The LCA results for Germany showed that it was possible to reduce German carbon dioxide emissions through storage and utilization to 640 Mtonne. This is a value lower than the target set by the European environmental policy for Germany. A sensitivity analysis showed that storage is more effective in reducing the value of GWP than additional utilization of carbon dioxide to produce useful products. Other impact categories remained constant or in some cases worsened.

The LCA results for Italy showed that total carbon emissions for Italy could be reduced to 249 Mtonne, a value below that required by the national environmental policies. A sensitivity analysis showed that the value of GWP was reduced if additional carbon dioxide was used to produce methane instead of being stored, keeping constant the overall quantity of CO_2 captured while other indicators of impact categories decreased or remained constant. This result suggests that, for the Italian CCUS supply chain, storage is less important to reduce the value of GWP and that a power to gas system has more beneficial results in this case. The power to gas system is predicted to be the most beneficial process to avoid an increase in the environmental impact. It is also a more mature technology.

This work shows how using LCA and sensitivity analysis helps find systems that increase the utilization rate of carbon dioxide while also reducting, or at least keeping constant, the GWP. The indicators for other impact categories are reduced only when the power to gas process is used for carbon dioxide utilization. Additional studies are recommended in order to develop more sustainable processes for power to gas to obtain a reduction of GWP even at high methane production rates.

Further developments are needed to improve the overall environmental burden of new carbon dioxide utilization routes in order to make them environmentally preferable to storage at higher utilization rates. Further improvement of conversion efficiencies would allow a wider choice from among the various carbon dioxide utilization options. This would contribute further to the reduction of emissions over the case when only methane production is the preferred route. This also agrees with the circular economy principles based on the recovery of a waste, in the case carbon dioxide, to produce different valuable products. In addition increasing carbon dioxide utilization options could reduce the overall cost by increasing revenues. More studies are needed to develop more environmentally friendly utilization routes. A trade-off between carbon dioxide storage and utilization is currently required and this needs thorough exploration in each case.

In a future study, it would be also interesting to analyze the same supply chains with different carbon dioxide sources, for example with carbon dioxide captured from ambient air.

Declarations

Not applicable

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References

Abotalib, M., Zhao, F., Clarens, A., 2016. Deployment of a Geographical Information System Life Cycle Assessment Integrated Framework for Exploring the Opportunities and Challenges of Enhanced Oil Recovery Using Industrial CO2 Supply in the United States, Sustainable Chem. Eng. 4:4743−4751.

Antonetti, E., Iaquaniello, G., Salladini, A., Spadaccini, L., Perathoner, S., Centi, G., 2017. Waste-to-Chemicals for a Circular Economy: The Case of Urea Production (Waste-to-Urea), ChemSusChem 10:912 – 920.

Azzolina, N.A., Hamling, J.A., Peck, W.D., Gorecki, C.D., Nakles, D.V., Melzer, L.S., 2017. A life cycle analysis of incremental oil produced via CO2 EOR, Energy Procedia 114:6588 – 6596.

Aresta, M., Galatola, M., 1999. Life cycle analysis applied to the assessment of the environmental impact of alternative synthetic processes. The dimethylcarbonate case: part 1, Jour of Clean. Prod. 7(3):181–193.

Bernier, E., Lavigne, C., 2013. Life cycle assessment of kraft lignin for polymer applications, Int Jour of Life Cyc Assess, 18:520–528.

Biernacki, P., Rother, T., Paul, W., Werner, P., Steinigeweg, S., 2018. Environmental impact of the excess electricity conversion into methanol, Jour of Clean. Prod. 191:87-98.

Biswas, W.K., Graham, J., Kelly, K., John, M.B., 2010. Global warming contributions from wheat, sheep meat and wool production in Victoria, Australia e a life cycle assessment, Jour of Clean. Prod. 18:1386-1392.

Biswas, W.K., Barton, L., Carter, D., 2008. Global warming potential of wheat production inWestern Australia: a life cycle assessment, Water Environ. J. 22:206–216.

Bouman, E.A., Ramirez, A., Hertwich, E.G., 2015. Multiregional environmental comparison of fossil fuel power generation—Assessment of the contribution of fugitive emissions from conventional and unconventional fossil resources, Int. J. Greenh. Gas Control, 33:1-9.

Boyer, M., Ponssard, J.P., 2013. Economic analysis of the European cement industry. 2013. Available at: https://hal.archives-ouvertes.fr/hal-00915646

Brentner, L., Eckelman, M., Zimmerman, J., 2011. Combinatorial life cycle assessment to inform process design of industrial production of algae biodiesel. Environ. Sci. Technol. 45:7060–7067.

Colbertaldo, P., Guandalini, G., Campanari, S., 2018. Modelling the integrated power and transport energy system: The role of power-to-gas and hydrogen in long-term scenarios for Italy, Energy. 154(1):592-601

Corsten, M., Ramirez, A., Shen, L., Koornneef, J., Faaij, A. 2013. Environmental impact assessment of CCS chains e lessons learned and limitations from LCA literature. Int J Greenh Gas Control 13:59-71.

Cooney, G., Littlefield, J., Marriott, J., Skone, T. 2015. Evaluating the climate benefits of CO2-enhanced oil recovery using life cycle analysis. Environ. Sci. Technol. 49:7491.

Cuéllar-Franca, R., Dimitriou, I., García-Gutierrez, P., Elder, R.H., Allen, R.W.K., Azapagic, A., 2015a. Carbon capture and utilisation: application of life cycle thinking to process design. Comp. Aid. Chem. Eng. 37:1457–1462.

Cuellar-Franca, R.M., Azapagic, A., 2015b. Carbon capture, storage and utilisation technologies: a critical analysis and comparison of their life cycle environmental impacts. J. CO2 Util. 9:82–102.

Erda, L., Wei, X., Hui, J., Yinlong, X., Yue, L., Liping, B., Liyong, X. 2005. Climate change impacts on crop yield and quality with CO2 fertilization in China, Phil. Trans. R. Soc. B, 360, 2149–2154

Europe, 2019. Available at: https://ec.europa.eu/eurostat/statistics-explained/index.php/Electricity_price_statistics(Accessed 14 March 2019).

European Commission, 2015a. Energy Union Package—A Framework Strategy for a Resilient Energy Union with a Forward-Looking Climate Change Policy, Brussels. Available at: https://www.eea.europa.eu/policy-documents/com-2015-80-final (accessed 4.5.2019)

European Commission, 2015b. Towards an Integrated Strategic Energy Technology (SET) Plan: Accelerating the European Energy System Transformation. Eurostat, 2016. ECU/EUR Exchane Rates Versus National Currencies – Code: Tec00033, Available at: http://ec.europa.eu/eurostat/tgm/table.do?tab=table&init=1&language=en&pcode=tec00033&plugin=1 (accessed 4.8.16).

Fernández-Dacostaa, C., van der Speka, M., Hung, C.R., Oregionni, G.D., Skagestad, R., Parihard, P., Gokakd, D.T., Strømmanb, A.H., Ramireza, A., 2017. Prospective techno-economic and environmental assessment of carbon capture at a refinery and CO2 utilisation in polyol synthesis, J. CO_2 Util. 21:405–422

Fernández-Dacosta, C., Stojcheva, V., Ramirez, A., 2018. Closing carbon cycles: Evaluating the performance of multi-product CO2 utilisation and storage configurations in a refinery, J. CO2 Util, 23:128-142.

Focus Economics, 2019. Available at: https://www.focus-economics.com/commodities/base-metals/steel-europe (Accessed 15.4.2019).

General Secretariat of the Council, 2014. Conclusions on 2030 climate and energy policy framework. Brussels. Available at : https://www.consilium.europa.eu/uedocs/cms_data/docs/pressdata/en/ec/145356.pdf (Accessed 4.5.2019)

Giordano, L., Roizard, D., Favre, E., 2018. Life cycle assessment of post-combustion CO2 capture: A comparison between membrane separation and chemical absorption processes, Int. J. Greenh. Gas Control 68:146–163.

Götz, M., Lefebvre, J., Mörs, F., McDaniel Koch, A., Graf, F., Bajohr, S., Kolb, R., 2016. Renewable Power-to-gas: A technological and economic review, Renew. Energy 85:1371–1390,

Gracceva, F., De Luca, E., Fidanza, A., Del Nero, P., Giuffrida, L.G., Felici, B., Pona, C., Zini, A., 2017. Analisi trimestrale del sistema energetico italiano. Report 1/2017. ISSN 2531-4750

Gursel, A.P., Horvath, A., 2012. Green Concrete LCA Tool. Univ. California, Berkeley, CA. Available at: https://greenconcrete.berkeley.edu/ (Accessed on 14.5.2019)

Guandalini, G., Robinius, M., Grube, T., Campanari, S., Stolten, D., 2017. Long-term power-to-gas potential from wind and solar power: A Country analysis for Italy, Int. J. of Hydrog. Energy. 42(19):13389-13406

Guinee, J.B., Gorree, M., Heijungs, R., Huppes, G., Kleijn, R., Sleeswijk, A.W., Haes, H., Au, D., Bruijn, J.A.D., Duin, R.V., Hujbregts, M.A.J., 2002. Handbook on Life Cycle Assessment: Operational Guide to the ISO Standards. Kluwer Academic Publisher, Dordrecht.

Han, J.H., Lee, I.B., 2013. A Comprehensive Infrastructure Assessment Model for Carbon Capture and Storage Responding to Climate Change under Uncertainty, Ind. Eng. Chem. Res. 52:3805−3815.

Hertwich, E.G., Aaberg, M., Singh, B., Strømman, A.H., 2008. Life-cycle assessment of carbon dioxide capture for enhanced oil recovery. Chin. J. Chem. Eng. 16(3):343–353.

Heijungs, R. 2014. Ten easy lessons for good communication of LCA. Int Jour of Life Cyc Assess 19:473–476

Hussain, D., Dzombak, D.A., Jaramillo, P., Lowry, G.V. 2013. Comparative lifecycle inventory (LCI) of greenhouse gas (GHG) emissions of enhanced oil recovery (EOR) methods using different CO2 sources. Int. J. Greenhouse Gas Control. 16:129−144.

Hornafius, K.Y., Hornafius, J.S., 2015. Carbon negative oil: A pathway for CO2 emission reduction goals. Int. J. Greenhouse Gas Control 37: 492−503.

IPCC, 2005, IPCC special report on Carbon dioxide Capture and Storage. Prepared by Working Group III of the Intergovernmental Panel on Climate Change [Metz, B., O. Davidson, H.C. de Coninck, M. Loos and L.A. Meyer (eds.)], Cambridge University Press, Cambridge, United Kingdom and New York, NY, USA, 442 pp.

ISO 14040. 2009, Environmental management – Life cycle assessment – Principles and framework, European Committee for Standardisation, Brussels, 2009.

ISO 14044. 2006, Environmental management – Life cycle assessment – Requirements and guidelines, European Committee for Standardisation, Brussels, 2006ISPRA. PBL and JRC, Trends in Global CO2 Emissions: 2013 Report, PBL Netherlands Environmental Assessment Agency and European Commission Joint Research Centre Institute for Environment and Sustainability, The Hague/Ispra (2013)

Jiang, Y., Lei, Y., Yang, Y., Wang, F., 2018. Life Cycle CO2 Emission Estimation of CCS-EOR System Using Different CO2 Sources, Pol. J. Environ. Stud. 27(6):2573-2583

Kahn, B., 2016. Available at: http://www.climatecentral.org/news/world-passes-400-ppmthreshold-permanently-20738. (accessed on 2.5.2019)

Kajaste, R., Hurme, M., and Oinas, P., 2018. Methanol-Managing greenhouse gas emissions in the production chain by optimizing the resource base, AIMS Energy, 6(6):1074–1102.

Khoo, Z.Y., Ho, EHZ, Li, Y, Yeo, Z, Low, JSC, Bu, J, Chia, LSO, 2021. Life cycle assessment of a CO2 mineralisation technology for carbon capture and utilisation in Singapore, Journal of CO2 Utilization 44, 101378

Kim, Y., Lim, S.R., Jung, K.A., Park, J.M. 2019. Process-based life cycle CO2 assessment of an ammonia-based carbon capture and storage system, Journal of Industrial and Engineering Chemistry 76, 223–232

Koornneef, J., van Keulen, T., Faaij, A., Turkenburg, W., 2008. Life cycle assessment of a pulverized coal power plant with post-combustion capture, transport and storage of CO2. Int. J. Greenh. Gas Control. 2:448-467.

Korre, A., Nie, Z., Durucan, S., 2010. Life cycle modelling of fossil fuel power generation with post-combustion CO2 capture. Int. J. Greenh. Gas Control. 4:289–300.

Leonzio, G., Foscolo, P.U. 2020. Analysis of a 2-D model of a packed bed reactor for methanol production by means of CO2 hydrogenation, International Journal of Hydrogen Energy. 45, 18, 10648-10663

Leonzio, G., Zondervan, E., Foscolo, P. U. 2019. Methanol production by CO2 hydrogenation: Analysis and simulation of reactor performance. International journal of hydrogen energy, 44(16), 7915-7933

Leonzio, G., Foscolo, P.U., Zondervan, E., 2019 Sustainable utilization and storage of carbon dioxide: analysis and design of an innovative supply chain, Comput. Chem. Eng., 135(5):106569.

Leonzio, G., Zondervan, E., 2020 Analysis and optimization of carbon supply chain integrated by power to gas process in Italy, Jour of Clean. Prod. 269:122172.

Liu, Y., Gea, J., Liu, C., He, R. 2020. Evaluating the energy consumption and air emissions of CO2-enhanced oil recovery in China: A partial life cycle assessment of extralow permeability reservoirs, International Journal of Greenhouse Gas Control 92, 102850

Mattila, H.P., Hudd, H., Zevenhoven, R., 2014. Cradle-to-gate life cycle assessment of precipitated calcium carbonate production from steel converter slag, Jour of Clean. Prod. 84:611-618.

Dowell, M., Fennell, N., Shah, P.S., Maitland NGC, (2017) The role of CO2 capture and utilization in mitigating climate change. Nat. Clim. Chang. 7:243–249.

Matzen, M., Demirel, Y., 2016a. Methanol and dimethyl ether from renewable hydrogen and carbon dioxide: Alternative fuels production and life-cycle assessment, Jour of Clean. Prod. 139, 1068-1077.

Meylana, F.D., Pigueta, F.P., Erkman, S., 2017. Power-to-gas through CO2 methanation: Assessment of the carbon balance regarding EU directives, J. Energy Storage 11, 16–24

Michailos, S., Sanderson, P., Zaragoza, A.V., McCord, S., Armstrong, K., Styring, P., 2018. Methanol Worked Examples for the TEA and LCA Guidelines for CO2 Utilization, Available at : https://deepblue.lib.umich.edu/handle/2027.42/145723 (accessed on 5.5.2019)

Modahl, I.S., Nyland, C.A., Raadal, H.L., Karstad, O., Torp, T.A., Hagemann, R., LCA as an ecodesign tool for production of electricity, including carbon capture and storage–A study of a gas power plant case with post-combustion CO2 capture at Tjeldbergodden. Proceedings of the Conference: Joint actions on climate change, Aalborg, Den, June 2009

Nikbin, I.M., Aliaghazadeh, M., Charkhta, S., Fathollahpour, A., 2018. Environmental impacts and mechanical properties of lightweight concrete containing bauxite residue (red mud), J. Clean. Prod. 172:2683-2694.

OECD, 2013. Effective Carbon Prices, Available at: http://www.oecd.org/env/tools-evaluation/carbon-prices.htm (Accessed 13 April 2019).

Ochoa Bique, A., Nguyen, T.B.H., Leonzio, G., Galanopoulos, C., Zondervan, E., 2018. Integration of carbon dioxide and hydrogen supply chains, Comput. Aided Chem. Eng., 43:1413-1418.

Pacala, S., Socolow, R., 2004. Stabilization wedges: solving the climate problem for the next 50 years with current technologies. Science 305(5686):968–972.

Parra, D., Zhang, X., Bauer, C., Patel, M.K., 2017. An integrated techno-economic and life cycle environmental assessment of power-to-gas systems, Appl. Energy 193:440–454.

Passell, H., Dhaliwal, H., Reno, M., Wu, B., Amotz, A.B., Ivry, E., Gay, M., Czartoski, T., Laurin, L., Ayer, N., 2013 Algae biodiesel life cycle assessment using current commercial data, J. Environ. Manage. 129:103–111.

Patricio, J., Angelis-Dimakis, A., Castillo-Castillo, A., Kalmykova, Y., Rosado, L., 2017. Region prioritization for the development of carbon capture and utilization technologies, J. CO2 Util. 17:50–59.

Pehnt, A., Henkel, J., 2009. Life cycle assessment of carbon dioxide capture and storage from lignite power plants. Int J Greenh Gas Control 3:49-66.

Petrescu, L., Bonalumi, D., Valenti, G., Cormos, A.M., Cormos, C.C., 2017. Life Cycle Assessment for supercritical pulverized coal power plants with post-combustion carbon capture and storage, Jour of Clean. Prod. 157:10-21.

Portdata, 2019 Available at https://www.pordata.pt/en/Europe/Electricity+prices+for+households+and+industrial+users+(Euro+ECU)-1477-211472 (Accessed 15.3.2019)

Reiter, G., Lindorfer, J., 2015. Global warming potential of hydrogen and methane production from renewable electricity via power-to-gas technology, Int Jour of Life Cyc Assess, 20:477–489

Rhodes, J., Clarens, A., Eranki, P., Long, J. 2015. Electricity from natural gas with CO2 capture for enhanced oil recovery; California Council on Science and Technology, ISBN 978-1-930117-98-3.

Singh, B., Strømman, A.H., Hertwich, E., 2011a. Life cycle assessment of natural gas combined cycle power plant with post-combustion carbon capture, transport and storage, Int. J. Greenhouse Gas Control 5(3):457–466.

Singh, B., Strømman, A.H., Hertwich, E.G., 2011b. Comparative life cycle environmental assessment of CCS technologies. Int J Greenhouse Gas Control 5(4):911–921.

Stangeland, K., Kalai, D., Li, H., Yu, Z., 2017. CO_2 methanation: the effect of catalysts and reaction conditions, Energy Procedia 105:2022 – 2027.

Štefanica, J., Smutná, J., Kočí, V., Machač, P., Pilař, L. 2016. Environmental gains and impacts of a CCS technology – case study of post-combustion CO2 separation by ammonia absorption, Energy Procedia 86:215–218

Sternberg, A., Bardow, A., 2016. Life Cycle Assessment of Power-to-Gas: Syngas vs Methane, ACS Sustainable Chem. Eng. 4:4156−4165.

Sternberg, A., and Bardow, A. 2015. Power-to-What? – Environmental assessment of energy storage systems, Energy Environ. Sci., 8:389–400.

Thinkstep,2019. Available at https://www.thinkstep.com/. (accessed on 4.5.2019)

Thorne, R.J., Sundsetha, K., Bouman, E., Czarnowska, L., Mathisend, A., Skagestadd, R., Stanekc, W., Pacynaa, J.M., Pacyna, E.G. 2020, Technical and environmental viability of a European CO2 EOR system, International Journal of Greenhouse Gas Control 92, 102857

von der Assen, N., and Bardow, A., 2014b. Life cycle assessment of polyols for polyurethane production using CO2 as feedstock: insights from an industrial case study, Green Chem., 16, 3272–3280.

von der Assen, N., Voll P., Peters, M., Bardow, A. 2014a. Life cycle assessment of CO2 capture and utilization: a tutorial review, Chem. Soc. Rev., 43:7982—7994.

von der Assen, N., Sternberg, A., Katelhon, A., Bardow, 2015. Environmental potential of carbon dioxide utilization in the polyurethane supply chain, Faraday Discuss., 183:291–307.

von der Aben, V.N., 2015. From Life-Cycle Assessment towards Life-Cycle Design of Carbon Dioxide Capture and Utilization, Available at: https://publications.rwth-aachen.de/record/570980/files/570980.pdf (Accessed on 4.5.2019)Volkart, K.,., Bauer, C., Boulet, C., 2013. Life cycle assessment of carbon capture and storage in power generation and industry in Europe, Int. J. Greenh. Gas Control 16:91–106.

Wildbolz, C., 2007. Life Cycle Assessment of Selected Technologies for CO2 Transport and Sequestration, Diploma Thesis, Available at: https://www.doka.ch/CCSDiplomaWildbolz07.pdf (accessed on 14.5.2019)

Yue, D., You, F., 2015. Integration of geological sequestration and microalgae biofixation supply chains for better greenhouse gas emission abatement, Chem. Eng. Trans., 45:487-492.

Zappa, W., 2014. Pilot-scale Experimental Work on the Production of Precipitated Calcium Carbonate (PCC) from Steel Slag for CO2 Fixation, Thesis submitted in partial fulfilment of the requirements for the degree of Master of Science in Technology Department of Energy Technology School of Engineering, Aalto University Espoo,

Zhang, X., Bauer, C., Mutel, C.L., Volkart, K., 2017. Life Cycle Assessment of Power-to-Gas: Approaches, system variations and their environmental implications, Appl. Energy 190:326–338.

Zhang, Z., Wang, T., Blunt, M. J., Anthony, E. J., Park, A-H. A., Hughes, R. W., Webley, P. A., & Yan, J. 2020. Advances in carbon capture, utilization and storage. Applied Energy, 278, 115627

Supplementary Materials

Life cycle assessment of a carbon capture utilization and storage supply chain in Italy and Germany: comparison between carbon dioxide storage and utilization systems

Grazia Leonzio*[1], I. David L. Bogle[2], Pier Ugo Foscolo[1]

[1]Department of Industrial and Information Engineering and Economics, University of L’Aquila, L’Aquila, 18 via G. Gronchi, 67100, Italy;

[2]Department of Chemical Engineering, Centre for Process Systems Engineering, University College London, London, Torrington Place WC1E 6BT, UK

*Corresponding author: grazia.leonzio@graduate.univaq.it

1. Life cycle inventory phase

Table S1

Results of LCIA analysis for the CCUS supply chain of Germany

AP	1.57×10^{9}	kgSO$_{2eq}$
EP	3.04×10^{10}	kgPhosphate$_{eq}$
ODP steady state	3.76×10^{3}	kgR11$_{eq}$
ADP elements	4.25×10^{5}	kgSb$_{eq}$
ADP fossil	3.44×10^{12}	MJ
FAETP	7.43×10^{10}	kgDCB$_{eq}$
HTP	5.27×10^{10}	kgDCB$_{eq}$
MAETP	4.54×10^{14}	kgDCB$_{eq}$
POCP	1.84×10^{8}	kgethene$_{eq}$
TETP	1.49×10^{9}	kgDCB$_{eq}$

Table S2

Results for different impact categories, considered in the LCIA analysis for the CCUS supply chain in Italy

AP	4.67×10^{8}	$kgSO_{2eq}$
EP	1.15×10^{11}	$kgPhosphate_{eq}$
ODP steady state	368	$kgR11_{eq}$
ADP elements	2.27×10^{4}	$kgSb_{eq}$
ADP fossil	1.96×10^{12}	MJ
FAETP	9.27×10^{10}	$kgDCB_{eq}$
MAETP	1.02×10^{13}	$kgDCB_{eq}$
POCP	7.88×10^{7}	$kgethene_{eq}$
TETP	4.42×10^{7}	$kgDCB_{eq}$

2. Results of sensitivity analysis

2.1 CCUS of Germany

Assuming that the fraction of carbon dioxide sent to the utilization section is used to produce methanol, and the captured carbon dioxide is stored at a rate of only 25%, 50%, 75% of the base case, the calculated methanol production is 9.8 Mton/year, 6.8 Mton/year and 3.8 Mton/year, respectively. Keeping constant the amount of captured carbon dioxide, and reducing the amount of carbon dioxide sent to storage (i.e. producing more methanol), the AP, EP, ODP, MAETP, POCP remain constant. On the other hand, the GWP, ADP elements and FAETP fossil increase compared to the base case. However, the GWP increases only slightly compared to base case. The opposite trend is observed for the ADP fossil, HTP and TETP. Overall, the variation of these impact categories is not significant compared to the base case.

In the following analysis, carbon dioxide that is not stored is utilized for concrete curing: when the stored amount of carbon dioxide is only 25%, 50%, 75% of the base case, CO_2-cured concrete is 513 Mton/year, 343 Mton/year and 174 Mton/year, respectively. Results show that for the ODP, ADP elements and MAETP no variations are present. The GWP, AP, EP, HTP, POCP, TETP are increased, then a higher environmental impact is present, especially for the POCP. The highest value that is achieved for the GWP is $3.46\cdot10^{11}$ $kgCO_{2\text{-}eq}$, when carbon dioxide stored is only 25% of the base case. Reductions are present for the FAETP and ADP fossil. Overall, producing a higher amount of CO_2-cured concrete increases the environmental impact.

In the following analysis, carbon dioxide that is not stored is used to produce urea. When stored carbon dioxide is only 25%, 50%, 75% of the base case, urea production is of 21.9 Mton/year, 15.5 Mton/year and 8.2 Mton/year, respectively. Results show that the TETP and MAETP have a constant trend, while the GWP, AP, EP, ADP fossil, HTP and POCP increase. The highest value for GWP is $2.28\cdot10^{11}$ $kgCO_{2\text{-}eq}$. On the other hand, only the FAETP is reduced increasing carbon dioxide sent to the utilization section. Overall, like in the previous case, the increase of urea production is not favorable to the reduction of the environmental impact.

Carbon dioxide that is not stored is sent to the utilization section to produce wheat. The total CO_2-assisted production of wheat, when carbon dioxide stored is only 25%, 50% and 75% of that sent to the storage section in the base case, is respectively $3.05 \cdot 10^{10}$ ton/year, $2.03 \cdot 10^{10}$ ton/year and $1.02 \cdot 10^{10}$ ton/year. With an increasing amount of carbon dioxide utilized for wheat production, only the GWP increases and a higher value compared to the base case is obtained ($8.55 \cdot 10^{12}$ $kgCO_{2\text{-eq}}$ compared to $1.94 \cdot 10^{11}$ $kgCO_{2\text{-eq}}$ of the base case). This suggests a higher environmental impact, even if a reduction of ADP fossil, FAETP, HTP and TETP is predicted. The other impact categories like POCP, MAETP, ADP elements, ODP, EP and AP present a constant trend.

Carbon dioxide not stored is sent to the utilization for lignin treatment: when only 25%, 50% and 75% of carbon dioxide sent to the storage section in the base case is stored, the respective amount of lignin that is upgraded is 69.7 Mton/year, 46.6 Mton/year and 23.5 Mton/year. Overall, increasing the lignin that is treated determines a higher environmental impact. In fact, the GWP, AP, ODP, ADP elements, ADP fossil, HTP, MAETP and POCP increase. However, as in the methanol case, no significant variations are obtained. For example, the highest value of GWP is $2.06 \cdot 10^{11}$ $kgCO_{2\text{-eq}}$. On the other hand, the TETP and FAETP show no variation.

When increasing the amount of carbon dioxide sent to the utilization for polyurethane production, it is evident that the environmental impact increases. The highest variation compared to the base case is present for the ADP elements, while for the other impact categories no significant variations compared to the base case are obtained. The highest GWP is $2.88 \cdot 10^{11}$ $kgCO_{2\text{-eq}}$. When only 25%, 50% and 75% of carbon dioxide sent to storage section in the base case is stored, the amount of polyurethane that is produced is respectively 62 Mton/year, 45 Mton/year and 28 Mton/year. Polyurethane is obtained in a conventional way by polyols and isocyanate, however it should be stressed here that we are not considering the traditional route for polyols production. These are obtained from carbon dioxide: CO_2 reacts with epoxides to produce polycarbonate polyols via a catalytic reaction (Orgilés-Calpena et al., 2016). The mechanical properties of resulting polyurethane are comparable with those obtained through a traditional way (Orgilés-Calpena et al., 2016).

In the following sensitivity analysis, the amount of carbon dioxide that is not stored is sent to the utilization for the production of calcium carbonate. When only 25%, 50%, 75% of carbon dioxide sent to the storage section in the base case is stored, the amount of calcium carbonate that is produced is respectively 135 Mton/year, 131 Mton/year and 126 Mton/year. A constant trend is present for the EP, ODP and FAETP. Increasing the utilization option, a reduction is obtained only for the ADP fossil, while an increment is obtained for other impact categories. The highest value for GWP is $1.96 \cdot 10^{11}$ $kgCO_{2\text{-eq}}$. However, no significant variations compared to base case are present.

In the last sensitivity analysis, carbon dioxide not stored is sent to utilization for the production of concrete by red mud. When only 25%, 50% and 75% of carbon dioxide sent to the storage in the base case is actually stored, the amount of concrete produced is respectively 1.16 billion ton/year, 783 Mton/year and 401 Mton/year. A constant trend is present for the ODP, ADP elements, FAETP and MAETP. Generally, increasing the amount of carbon dioxide sent to the utilization section, the GWP, AP, EP, HTP, POCP and TETP increase, while only the ADP fossil decreases. The highest value of GWP is $5.79\cdot10^{11}$ kg$CO_{2\text{-eq}}$, calculated when only 25% of carbon dioxide is stored in storage section compared to the base case. However, no substantial variations are predicted compared to base case for these impact categories.

The following Tables S3 – S10 summarize the results obtained with the sensitivity analysis for the CCUS of Germany, with reference to different scenarios (see also the methodology applied in Xiang et al. (2015)

Table S3 Results of sensitivity analysis considering methanol production for the CCUS of Germany

	Base case (A)	25% CO_2 stored compared A	50% CO_2 stored compared A	75% CO_2 stored compared A
GWP (kgCO_{2eq})	1.94×10^{11}	1.98×10^{11}	1.97×10^{11}	1.95×10^{11}
AP (kgSO_{2eq})	1.57×10^{9}	1.57×10^{9}	1.57×10^{9}	1.57×10^{9}
EP (kgPhoshate$_{eq}$)	3.04×10^{10}	3.04×10^{10}	3.04×10^{10}	3.04×10^{10}
ODP (kgR11$_{eq}$)	3.76×10^{3}	3.76×10^{3}	3.76×10^{3}	3.76×10^{3}
ADP elements (kgSb$_{eq}$)	4.25×10^{5}	4.28×10^{5}	4.27×10^{5}	4.26×10^{5}
ADP fossil (MJ)	3.44×10^{12}	3.39×10^{12}	3.40×10^{12}	3.42×10^{12}
FAETP (kgDCB$_{eq}$)	7.43×10^{10}	7.48×10^{10}	7.46×10^{10}	7.44×10^{10}
HTP inf (kgDCB$_{eq}$)	5.27×10^{10}	5.25×10^{10}	5.26×10^{10}	5.26×10^{10}
MAETP (kgDCB$_{eq}$)	4.54×10^{14}	4.54×10^{14}	4.54×10^{14}	4.54×10^{14}
POCP (kgethene$_{eq}$)	1.84×10^{8}	1.84×10^{8}	1.84×10^{8}	1.84×10^{8}
TETP (kgDCB$_{eq}$)	1.49×10^{9}	1.48×10^{9}	1.49×10^{9}	1.49×10^{9}

Table S4 Results of sensitivity analysis considering concrete curing for the CCUS of Germany

	Base case (A)	25% CO_2 stored compared A	50% CO_2 stored compared A	75% CO_2 stored compared A
GWP (kgCO_{2eq})	1.94×10^{11}	3.46×10^{11}	2.95×10^{11}	2.45×10^{11}
AP (kgSO_{2eq})	1.57×10^{9}	2.43×10^{9}	2.15×10^{9}	1.86×10^{9}
EP (kgPhoshate$_{eq}$)	3.04×10^{10}	3.05×10^{10}	3.05×10^{10}	3.04×10^{10}
ODP (kgR11$_{eq}$)	3.76×10^{3}	3.76×10^{3}	3.76×10^{3}	3.76×10^{3}
ADP elements (kgSb$_{eq}$)	4.25×10^{5}	4.25×10^{5}	4.25×10^{5}	4.25×10^{5}
ADP fossil (MJ)	3.44×10^{12}	3.38×10^{12}	3.40×10^{12}	3.42×10^{12}
FAETP (kgDCB$_{eq}$)	7.43×10^{10}	7.42×10^{10}	7.42×10^{10}	7.42×10^{10}
HTP inf (kgDCB$_{eq}$)	5.27×10^{10}	5.56×10^{10}	5.46×10^{10}	5.37×10^{10}
MAETP (kgDCB$_{eq}$)	4.54×10^{14}	4.54×10^{14}	4.54×10^{14}	4.54×10^{14}
POCP (kgethene $_{eq}$)	1.84×10^{8}	1.12×10^{9}	8.11×10^{8}	4.98×10^{8}
TETP (kgDCB$_{eq}$)	1.49×10^{9}	1.57×10^{9}	1.54×10^{9}	1.52×10^{9}

Table S5 Results of sensitivity analysis considering urea production for the CCUS of Germany

	Base case (A)	25% CO$_2$ stored compared A	50% CO$_2$ stored compared A	75% CO$_2$ stored compared A
GWP (kgCO$_{2eq}$)	1.94×10^{11}	2.28×10^{11}	2.16×10^{11}	2.05×10^{11}
AP (kgSO$_{2eq}$)	1.57×10^{9}	1.72×10^{9}	1.67×10^{9}	1.62×10^{9}
EP (kgPhoshate$_{eq}$)	3.04×10^{10}	3.07×10^{10}	3.06×10^{10}	3.05×10^{10}
ODP (kgR11$_{eq}$)	3.76×10^{3}	3.77×10^{3}	3.76×10^{3}	3.76×10^{3}
ADP elements (kgSb$_{eq}$)	4.25×10^{5}	4.28×10^{5}	4.27×10^{5}	4.26×10^{5}
ADP fossil (MJ)	3.44×10^{12}	3.80×10^{12}	3.68×10^{12}	3.56×10^{12}
FAETP (kgDCB$_{eq}$)	7.43×10^{10}	7.42×10^{10}	7.42×10^{10}	7.42×10^{10}
HTP inf (kgDCB$_{eq}$)	5.27×10^{10}	5.28×10^{10}	5.28×10^{10}	5.27×10^{10}
MAETP (kgDCB$_{eq}$)	4.54×10^{14}	4.54×10^{14}	4.54×10^{14}	4.54×10^{14}
POCP (kgethene $_{eq}$)	1.84×10^{8}	1.86×10^{8}	1.85×10^{8}	1.85×10^{8}
TETP (kgDCB$_{eq}$)	1.49×10^{9}	1.49×10^{9}	1.49×10^{9}	1.49×10^{9}

Table S6 Results of sensitivity analysis considering wheat production for the CCUS of Germany

	Base case (A)	25% CO$_2$ stored compared A	50% CO$_2$ stored compared A	75% CO$_2$ stored compared A
GWP (kgCO$_{2eq}$)	1.94×10^{11}	8.55×10^{12}	5.77×10^{12}	2.98×10^{12}
AP (kgSO$_{2eq}$)	1.57×10^{9}	1.57×10^{9}	1.57×10^{9}	1.57×10^{9}
EP (kgPhoshate$_{eq}$)	3.04×10^{10}	3.04×10^{10}	3.04×10^{10}	3.04×10^{10}
ODP (kgR11eq)	3.76×10^{3}	3.76×10^{3}	3.76×10^{3}	3.76×10^{3}
ADP elements (kgSb$_{eq}$)	4.25×10^{5}	4.25×10^{5}	4.25×10^{5}	4.25×10^{5}
ADP fossil (MJ)	3.44×10^{12}	3.38×10^{12}	3.40×10^{12}	3.42×10^{12}
FAETP (kgDCB$_{eq}$)	7.43×10^{10}	7.42×10^{10}	7.42×10^{10}	7.42×10^{10}
HTP inf (kgDCB$_{eq}$)	5.27×10^{10}	5.25×10^{10}	5.26×10^{10}	5.26×10^{10}
MAETP (kgDCB$_{eq}$)	4.54×10^{14}	4.54×10^{14}	4.54×10^{14}	4.54×10^{14}
POCP (kgethene $_{eq}$)	1.84×10^{8}	1.84×10^{8}	1.84×10^{8}	1.84×10^{8}
TETP (kgDCB$_{eq}$)	1.49×10^{9}	1.48×10^{9}	1.49×10^{9}	1.49×10^{9}

Table S7 Results of sensitivity analysis considering lignin production for the CCUS of Germany

	Base case (A)	25% CO_2 stored compared A	50% CO_2 stored compared A	75% CO_2 stored compared A
GWP (kgCO_{2eq})	1.94×10^{11}	2.06×10^{11}	2.02×10^{11}	1.98×10^{11}
AP (kgSO_{2eq})	1.57×10^{9}	1.64×10^{9}	1.62×10^{9}	1.60×10^{9}
EP (kgPhoshate$_{eq}$)	3.04×10^{10}	3.04×10^{10}	3.04×10^{10}	3.04×10^{10}
ODP (kgR11$_{eq}$)	3.76×10^{3}	3.77×10^{3}	3.77×10^{3}	3.76×10^{3}
ADP elements (kgSb$_{eq}$)	4.25×10^{5}	5.18×10^{5}	4.87×10^{5}	4.56×10^{5}
ADP fossil (MJ)	3.44×10^{12}	4.96×10^{12}	4.45×10^{12}	3.95×10^{12}
FAETP (kgDCB$_{eq}$)	7.43×10^{10}	7.43×10^{10}	7.43×10^{10}	7.43×10^{10}
HTP inf (kgDCB$_{eq}$)	5.27×10^{10}	5.30×10^{10}	5.29×10^{10}	5.28×10^{10}
MAETP (kgDCB$_{eq}$)	4.54×10^{14}	4.55×10^{14}	4.54×10^{14}	4.54×10^{14}
POCP (kgethene $_{eq}$)	1.84×10^{8}	1.88×10^{8}	1.86×10^{8}	1.85×10^{8}
TETP (kgDCB$_{eq}$)	1.49×10^{9}	1.49×10^{9}	1.49×10^{9}	1.49×10^{9}

Table S8 Results of sensitivity analysis considering polyurethane production for the CCUS of Germany

	Base case (A)	25% CO_2 stored compared A	50% CO_2 stored compared A	75% CO_2 stored compared A
GWP (kgCO_{2eq})	1.94×10^{11}	2.88×10^{11}	2.56×10^{11}	2.20×10^{11}
AP (kgSO_{2eq})	1.57×10^{9}	1.78×10^{9}	1.71×10^{9}	1.64×10^{9}
EP (kgPhoshate$_{eq}$)	3.04×10^{10}	3.05×10^{10}	3.05×10^{10}	3.04×10^{10}
ODP (kgR11$_{eq}$)	3.76×10^{3}	3.96×10^{3}	3.89×10^{3}	3.82×10^{3}
ADP elements (kgSb$_{eq}$)	4.25×10^{5}	1.50×10^{6}	1.14×10^{6}	7.81×10^{5}
ADP fossil (MJ)	3.44×10^{12}	5.52×10^{12}	4.83×10^{12}	4.13×10^{12}
FAETP (kgDCB$_{eq}$)	7.43×10^{10}	7.49×10^{10}	7.46×10^{10}	7.44×10^{10}
HTP inf (kgDCB$_{eq}$)	5.27×10^{10}	8.12×10^{10}	7.17×10^{10}	6.22×10^{10}
MAETP (kgDCB$_{eq}$)	4.54×10^{14}	4.60×10^{14}	4.58×10^{14}	4.56×10^{14}
POCP (kgethene $_{eq}$)	1.84×10^{8}	2.07×10^{8}	1.99×10^{8}	1.91×10^{8}
TETP (kgDCB$_{eq}$)	1.49×10^{9}	1.57×10^{9}	1.54×10^{9}	1.52×10^{9}

Table S9 Results of sensitivity analysis considering calcium carbonate production for the CCUS of Germany

	Base case (A)	25% CO_2 stored compared A	50% CO_2 stored compared A	75% CO_2 stored compared A
GWP (kgCO_{2eq})	1.94×10^{11}	1.96×10^{11}	1.96×10^{11}	1.95×10^{11}
AP (kgSO_{2eq})	1.57×10^{9}	1.67×10^{9}	1.63×10^{9}	1.60×10^{9}
EP (kgPhoshate$_{eq}$)	3.04×10^{10}	3.04×10^{10}	3.04×10^{10}	3.04×10^{10}
ODP (kgR11$_{eq}$)	3.76×10^{3}	3.76×10^{3}	3.76×10^{3}	3.76×10^{3}
ADP elements (kgSb$_{eq}$)	4.25×10^{5}	4.30×10^{5}	4.28×10^{5}	4.27×10^{5}
ADP fossil (MJ)	3.44×10^{12}	3.40×10^{12}	3.41×10^{12}	3.43×10^{12}
FAETP (kgDCB$_{eq}$)	7.43×10^{10}	7.43×10^{10}	7.43×10^{10}	7.43×10^{10}
HTP inf (kgDCB$_{eq}$)	5.27×10^{10}	5.65×10^{10}	5.52×10^{10}	5.40×10^{10}
MAETP (kgDCB$_{eq}$)	4.54×10^{14}	5.03×10^{14}	4.87×10^{14}	4.71×10^{14}
POCP (kgethene $_{eq}$)	1.84×10^{8}	1.90×10^{8}	1.88×10^{8}	1.86×10^{8}
TETP (kgDCB$_{eq}$)	1.49×10^{9}	1.63×10^{9}	1.59×10^{9}	1.54×10^{9}

Table S10 Results of sensitivity analysis considering concrete production from red mud for the CCUS of Germany

	Base case (A)	25% CO_2 stored compared A	50% CO_2 stored compared A	75% CO_2 stored compared A
GWP (kgCO_{2eq})	1.94×10^{11}	5.79×10^{11}	4.51×10^{11}	3.22×10^{11}
AP (kgSO_{2eq})	1.57×10^{9}	3.05×10^{9}	2.56×10^{9}	2.06×10^{9}
EP (kgPhoshate$_{eq}$)	3.04×10^{10}	3.05×10^{10}	3.05×10^{10}	3.05×10^{10}
ODP (kgR11$_{eq}$)	3.76×10^{3}	3.76×10^{3}	3.76×10^{3}	3.76×10^{3}
ADP elements (kgSb$_{eq}$)	4.25×10^{5}	4.25×10^{5}	4.25×10^{5}	4.25×10^{5}
ADP fossil (MJ)	3.44×10^{12}	3.38×10^{12}	3.40×10^{12}	3.42×10^{12}
FAETP (kgDCB$_{eq}$)	7.43×10^{10}	7.43×10^{10}	7.43×10^{10}	7.43×10^{10}
HTP inf (kgDCB$_{eq}$)	5.27×10^{10}	5.91×10^{10}	5.69×10^{10}	5.48×10^{10}
MAETP (kgDCB$_{eq}$)	4.54×10^{14}	4.54×10^{14}	4.54×10^{14}	4.54×10^{14}
POCP (kgethene $_{eq}$)	1.84×10^{8}	1.86×10^{9}	1.30×10^{9}	7.41×10^{8}
TETP (kgDCB$_{eq}$)	1.49×10^{9}	1.67×10^{9}	1.61×10^{9}	1.55×10^{9}

Table S11 Results of sensitivity analysis for methanol production with an efficiency of 90%

	Base case (A)	25% CO_2 stored compared A	50% CO_2 stored compared A	75% CO_2 stored compared A
GWP ($kgCO_{2eq}$)	1.94×10^{11}	1.94×10^{11}	1.94×10^{11}	1.94×10^{11}
AP ($kgSO_{2eq}$)	1.57×10^{9}	1.57×10^{9}	1.57×10^{9}	1.57×10^{9}
EP ($kgPhoshate_{eq}$)	3.04×10^{10}	3.04×10^{10}	3.04×10^{10}	3.04×10^{10}
ODP ($kgR11_{eq}$)	3.76×10^{3}	3.76×10^{3}	3.76×10^{3}	3.76×10^{3}
ADP elements ($kgSb_{eq}$)	4.25×10^{5}	4.28×10^{5}	4.27×10^{5}	4.26×10^{5}
ADP fossil (MJ)	3.44×10^{12}	3.39×10^{12}	3.40×10^{12}	3.42×10^{12}
FAETP ($kgDCB_{eq}$)	7.43×10^{10}	7.48×10^{10}	7.46×10^{10}	7.44×10^{10}
HTP inf ($kgDCB_{eq}$)	5.27×10^{10}	5.25×10^{10}	5.26×10^{10}	5.26×10^{10}
MAETP ($kgDCB_{eq}$)	4.54×10^{14}	4.54×10^{14}	4.54×10^{14}	4.54×10^{14}
POCP ($kgethene_{eq}$)	1.84×10^{8}	1.84×10^{8}	1.84×10^{8}	1.84×10^{8}
TETP ($kgDCB_{eq}$)	1.49×10^{9}	1.48×10^{9}	1.49×10^{9}	1.49×10^{9}

Table S12 Results of sensitivity analysis for methanol production with an efficiency of 100%

	Base case (A)	25% CO_2 stored compared A	50% CO_2 stored compared A	75% CO_2 stored compared A
GWP ($kgCO_{2eq}$)	1.94×10^{11}	1.94×10^{11}	1.94×10^{11}	1.94×10^{11}
AP ($kgSO_{2eq}$)	1.57×10^{9}	1.57×10^{9}	1.57×10^{9}	1.57×10^{9}
EP ($kgPhoshate_{eq}$)	3.04×10^{10}	3.04×10^{10}	3.04×10^{10}	3.04×10^{10}
ODP ($kgR11_{eq}$)	3.76×10^{10}	3.76×10^{3}	3.76×10^{3}	3.76×10^{3}
ADP elements ($kgSb_{eq}$)	4.25×10^{5}	4.28×10^{5}	4.27×10^{5}	4.26×10^{5}
ADP fossil (MJ)	3.44×10^{12}	3.39×10^{12}	3.40×10^{12}	3.42×10^{12}
FAETP ($kgDCB_{eq}$)	7.43×10^{10}	7.48×10^{10}	7.46×10^{10}	7.44×10^{10}
HTP inf ($kgDCB_{eq}$)	5.27×10^{10}	5.25×10^{10}	5.26×10^{10}	5.26×10^{10}
MAETP ($kgDCB_{eq}$)	4.54×10^{14}	4.54×10^{14}	4.54×10^{14}	4.54×10^{14}
POCP ($kgethene_{eq}$)	1.84×10^{8}	1.84×10^{8}	1.84×10^{8}	1.84×10^{8}
TETP ($kgDCB_{eq}$)	1.49×10^{9}	1.48×10^{9}	1.49×10^{9}	1.49×10^{9}

2.2 CCUS of Italy

Table S13 Results of sensitivity analysis for the CCUS of Italy

	Base case (A)	25% CO_2 stored compared A	50% CO_2 stored compared A	75% CO_2 stored compared A
GWP ($kgCO_{2eq}$)	9.62×10^{10}	9.24×10^{10}	9.39×10^{10}	9.48×10^{10}
AP ($kgSO_{2eq}$)	4.67×10^{8}	4.62×10^{8}	4.64×10^{8}	4.65×10^{8}
EP ($kgPhoshate_{eq}$)	1.15×10^{11}	1.15×10^{11}	1.15×10^{11}	1.15×10^{11}
ODP ($kgR11_{eq}$)	3.68×10^{2}	3.41×10^{2}	3.51×10^{2}	3.59×10^{2}
ADP elements ($kgSb_{eq}$)	2.27×10^{4}	2.23×10^{4}	2.24×10^{4}	2.26×10^{4}
ADP fossil (MJ)	1.96×10^{12}	1.91×10^{12}	1.93×10^{12}	1.94×10^{12}
FAETP ($kgDCB_{eq}$)	9.27×10^{10}	9.17×10^{10}	9.21×10^{10}	9.22×10^{10}
MAETP ($kgDCB_{eq}$)	1.02×10^{13}	9.64×10^{13}	9.84×10^{13}	1.00×10^{13}
POCP (kgethene $_{eq}$)	7.88×10^{7}	7.78×10^{7}	7.82×10^{7}	7.84×10^{7}
TETP ($kgDCB_{eq}$)	4.42×10^{7}	4.08×10^{7}	4.19×10^{7}	4.30×10^{7}

3. Description of processes for CO_2 utilization

3.1 Concrete curing

In the concrete curing process, carbon dioxide is injected into the curing vessels at room temperature and here diffuses into the fresh concrete under low pressure to produce calcium carbonate ($CaCO_3$). In this reaction, carbon dioxide reacts with cement components or hydration products such as $3CaO{\cdot}SiO_2$, $2CaO{\cdot}SiO_2$, $Ca(OH)_2$, $xCaO{\cdot}SiO_2{\cdot}yH_2O$ gel etc (Thomas Concrete, 2018; Xuang et al., 2018). After a few hours, the so obtained concrete has a higher compressive strength, better abrasion resistance, lower drying shrinkage and costs due to a reduction of cement content than in conventional concrete (Shi-Cong et al., 2014).

3.2 Wheat production

Carbon dioxide influences the photosynthesis, improving it because of its higher concentration in the surrounding atmosphere. However, an excess of carbon dioxide alters carbon (C) and nitrogen (N) metabolism, changing the chemical composition of agricultural plants (Hogy et al., 2009). This could determine higher yield but lower quality. the results of free air concentration enrichment (FACE) tests are sometimes contradicting the laboratory experiments about the quality (regarding the nitrogen and protein content) of the agricultural products (Nuttall et al. 2017; Verrillo et al., 2017). Generally, it is recommended to keep carbon dioxide concentration level just above that in the atmosphere in the growing environment where wheat is cultivated on large scale (Watson et al., 2018, Erda et al. doi:10.1098/rstb.2005.1743).

3.3 Lignin treatment

Lignin is obtained through the extraction of black liquor, from the pulp mill industry. In this condition, it is characterized by a pH between 13-14. However, to be used as raw material, a pH of about 8 should be achieved treating lignin with carbon dioxide (Patricio et al., 2017). The treated lignin can be used as an additive for concrete mixtures (Yufang et al., 2016), catalysts (Atul et al., 2013), polyethylene (Samal et al., 2014), propylene (Gregorová et al., 2005) and other chemicals.

3.4 Polyurethane production

Polyurethane is obtained in a conventional way by polyol and isocyanate through a catalytic reaction (von der Assen et al., 2015). These two reagents are petroleum derived products. However, an alternative to this conventional route is taken into account producing polyol from carbon dioxide. CO_2 reacts with epoxides to produce polycarbonate polyols via a catalytic reaction (Orgilés-Calpena et al., 2016). The mechanical properties of polyurethane are comparable with those obtained through a traditional way (Orgilés-Calpena et al., 2016).

3.5 Calcium carbonate production via mineral carbonation

Calcium carbonate is naturally produced and it is known as ground calcium carbonate (GCC). However, it can be industrially produced via precipitation and it is known as precipitated calcium carbonate (PCC). In this second route, steel slags are used as raw material that reacts with carbon dioxide to produce calcium carbonate (Lee et al., 2016). In fact, steel slags are mainly composed by CaO and MgO in addition to heavy metals as Mn, V, Zn, Cu, Ni, Cd, Pb, Sb, Mo, and Cr (Yadav and Mehra, 2017). An advantage of this process is that it can be controlled to have the desired quality, purity and size of crystals (Eloneva et al., 2008).

3.6 Urea production

Urea is obtained from the reaction of ammonia and carbon dioxide. Ammonia is produced by the reaction of hydrogen and nitrogen, where the first one is obtained by syngas obtained from natural gas reforming. In particular, two different steps are involved: at first ammonium carbamate is obtained in the liquid state while in the second step urea is formed by dehydrogenation of ammonium carbamate. Different process schemes are proposed by Koohestanian et al. (2018) and Edrisi et al. (2013, 2014a, 2014b, 2016).

3.7 Methanol production

A traditional way to produce methanol is the indirect way, via syngas hydrogenation, where syngas is obtained by the steam reforming of natural gas (Olah et al., 2005). A more environmentally friendly way is according to the catalytic direct hydrogenation of carbon dioxide using $CuO/ZnO/Al_2O_3$ as catalyst (Leonzio, 2018). Hydrogen can be obtained from the electrolysis of water exploiting renewable energies (solar or wind energies), from biomass pyrolysis, coke oven gas, reforming of biomass-derived products or partial oxidation of light oil residues (Leonzio, 2018). The hydrogenation of carbon dioxide is kinetically and thermodynamically limited then the recycle of unconverted gases after the separation of methanol and water, the utilization of membrane permeable to water are solutions that can be considered to improve conversions and yields (Leonzio et al., 2019).

3.8 Concrete by red mud production

Red mud, known also as "bauxite residue", is obtained by bauxite treatment in the alumina production. It is characterized by an high value of pH (between 10.5-12.5) due to the presence of Al_2O_3, Fe_2O_3, SiO_2, TiO_2, CaO, Na_2O, then it is disposed in landfills (Patricio et al., 2017). A way to reduce the pH and use it as a raw material consists on treating red mud with carbon dioxide. Generally, the treated read mud can be used as additive for building materials, as adsorbent for the removal of heavy metals, for the preparation of catalysts, ceramics, pigments, polymers and paints, for the recovery of iron, aluminum, titanium (Sutar et al., 2014; Liu et al., 2009). It is found that corrosion resistance, compressive strength, elasticity modulus, splitting tensile strength can be improved if concrete is composed by about 20% wt of red mud (Ribeiro et al., 2012; Liu and Poon, 2016).

3.9 Methane production

Methane can be produced by hydrogenation of carbon dioxide via power to gas system (Leonzio, 2017). In this case hydrogen is obtained via water electrolysis using fluctuant renewable energies. The hydrogenation reaction is called Sabatier reaction: it is exothermic and is carried out in a rage of temperature between 200 °C and 500 °C and at relatively high pressure (10-30 bar) (Stangeland et al., 2015). A Nickel based catalyst is used for this reaction, even if Ru, Rh and Co on various oxide supports (TiO_2, SiO_2, MgO, and Al_2O_3) can also be used (Brooks et al., 2007; Kopyscinski et al., 2010). Adiabatic fixed bed methanation reactors, isothermal fluidized bed reactors are used for this reaction (Di Felice and Micheli, 2015). The biological methanation can be also considered for power to gas systems (Ma et al., 2018).

References

Atul, M.V., Ekhe, J.D., 2013. Pyrolysis of purified kraft lignin in the presence of AlCl3 and ZnCl2, J. Environ. Chem. Eng. 1 (4):844–849

Brooks, K.P., Hu, J., Zhu, H., Kee, R.J., 2007. Methanation of carbon dioxide by hydrogen reduction using the Sabatier process in microchannel reactors, Chemical Engineering Science 62:1161 – 1170.

Di Felice, L., Micheli, F., 2015. From Biomass to SNG, Process Intensification for Sustainable Energy Conversion, First Edition. Edited by Fausto Gallucci and Martin van Sint Annaland. John Wiley & Sons, Ltd.

Edrisi, A., Mansoori, Z., Dabir, B., 2016. Urea synthesis using chemical looping process – Techno-economicevaluation of a novel plant configuration for a green production, International Journal of Greenhouse Gas Control 44 42–51.

Edrisi, A., 2013. Investigation of the chemical-looping systems for hydrogenproduction with inherent CO2separation and feasibility study of using this system in urea production process, Chemical Engineering. AmirkabirUniversity of Technology (Tehran Polytechnic).

Edrisi, A., Mansoori, Z., Dabir, B., 2014a. Using three chemical looping reactors inammonia production process – a novel plant configuration for a greenproduction. Int. J. Hydrogen Energy 39, 8271–8282.

Edrisi, A., Mansoori, Z., Dabir, B., Shahnazari, A., 2014b. Hydrogen, nitrogen andcarbon dioxide production through chemical looping using iron-based oxygencarrier – A Green plant for H2and N2production. Int. J. Hydrogen Energy.

Eloneva, S., Teir, S., Salminen, J., Fogelholm, C.J., Zevenhoven, R., 2008. Steel Converter Slag as a Raw Material for Precipitation of Pure Calcium Carbonate, Ind. Eng. Chem. Res. 47, 7104–7111

Erda, L., Wei, X., Hui, J., Yinlong, X., Yue, L., Liping, B., Liyong, X. 2005. Climate change impacts on crop yield and quality with CO2 fertilization in China, Phil. Trans. R. Soc. B, 360, 2149–2154

Gregorová, A., Cibulková, Z., Košíková, B., Šimon, P. 2005. Stabilization effect of lignin in polypropylene and recycled polypropylene, Polym. Degrad. Stab. 89 553–558

Hogy, P., Wieser, H., Kohler, P., Schwadorf, K., Breuer, J., Franzaring, J., Muntifering R., Fangmeier, A. 2009. Effects of elevated CO2 on grain yield and quality of wheat: results from a 3-year free-air CO2 enrichment experiment, Plant Biology ISSN 1435-8603.

Koohestanian, E., Sadeghi, J., Mohebbi-Kalhori, D., Shahraki, F., Samimi, A., 2018. A novel process for CO2 capture from the flue gases to produce urea and ammonia, Energy 144 279-285

Kopyscinski, J., Schildhauer, T.J., Biollaz, S.M.A., 2010. Production of synthetic natural gas (SNG) from coal and dry biomass –A technology review from 1950 to 2009, Fuel 89, 1763–1783.

Lee, S., Kim, J.W., Chae, S., Bang, J.H., Lee, S.W. 2016. CO2 sequestration technology through mineral carbonation: An extraction and carbonation of blast slag, Journal of CO2 Utilization 16 336–345

Leonzio, G., 2018. State of art and perspectives about the production of methanol, dimethyl ether and syngas by carbon dioxide hydrogenation. Journal of CO_2 Utilization 27 326–354

Leonzio, G., E. Zondervan, P.U. Foscolo, 2019. Methanol production by CO2 hydrogenation: Analysis and simulation of reactor performance, International Journal of Hydrogen Energy, 44, 16, 7915-7933

Leonzio, G., 2017. Design and feasibility analysis of Power-to-gas plant in Germany. Journal of Cleaner Production. 162, 609-623.

Liu, W.C., Yang, J.K., Xiao, B., 2009. Application of Bayer red mud for iron recovery and building material production from alumosilicate residues. J. Hazard. Mater. 161, 474-478.

Liu, R.X., Poon, C.S., 2016. Utilization of red mud derived from bauxite in self-compacting concrete, Journal of Cleaner Production 112 384-391

Long, S.P., Ainsworth E.A., Leakey A.D.B., Nosberger J., Ort D.R. 2006. Food for thought: lower-than-expected crop yield stimulation with rising CO2 concentrations. Science, 312, 1918–1921.

Ma, J., Li, Q., Kühn, M., Nakaten, N., 2018. Power-to-gas based subsurface energy storage: A review, Renewable and Sustainable Energy Reviews 97, 478–496.

Nuttall, J.G., O'Leary, G.J., Panozzo, J.F., Walkea, C.K., Barlow, K.M., Fitzgerald, G.J. 2017 Models of grain quality in wheat—Areview. Field Crops Research 202, 136–145.

Olah, G.A. 2005. Beyond oil and gas: the methanol economy, Angew. Chem. Int. Ed. 44 2636–2639

Orgilés-Calpena, E., Arán-Aís, F., Torró-Palau, A., Montiel-Parreño, E., Orgilés-Barceló, C. 2016. Synthesis of polyurethanes from CO2-based polyols: A challenge for sustainable adhesives, International Journal of Adhesion&Adhesives 67 63–68

Patricio, J., Angelis-Dimakis, A., Castillo-Castillo, A., Kalmykova, Y., Rosado, L. 2017. Region prioritization for the development of carbon capture and utilization technologies, Journal of CO2 Utilization 17 50–59.

Ribeiro, D.V., Labrincha, J.A., Morelli, M.R., 2012. Effect of the addition of red mud on the corrosion parameters of reinforced concrete. Cem. Concr. Res. 42, 124e133.

Sangram, K., Samal, E. G. Fernandes, Andrea Corti, Emo Chiellini, 2014. Bio-based Polyethylene–Lignin Composites Containing a Pro-oxidant/Pro-degradant Additive: Preparation and Characterization. J Polym Environ, 22:58–68

Shi-Cong, K., Bao-jian, Z., Chi-Sun, P., 2014. Use of a CO2 curing step to improve the properties of concrete prepared with recycled aggregates, Cement & Concrete Composites 45, 22–28.

Stangeland, K., Kalai, D., Li, H., Yu, Z., 2017. CO_2 methanation: the effect of catalysts and reaction conditions, Energy Procedia 105, 2022 – 2027.

Sutar, H., Mishra, S.C., Sahoo, S.K., Chakraverty, A.P., Maharana, H.S. 2014. Progress of Red Mud Utilization: An Overview, American Chemical Science Journal 4(3) 255-279

Thomas Concrete, 2018 available: http://thomasconcrete.com/images/pdf/Sustainability/Thomas-Concrete_CarbonCure_2018.pdf

Verrillo, F., Badeck, F.W., Terzi, V., Rizza, F., Bernardo, L., Di Maro, A., Fares, C., Zaldei, A., Miglietta, F., Moschella, A., Bracale, M., and Vannini, C., 2017. Elevated field atmospheric CO2 concentrations affect the characteristics of winter wheat (cv. Bologna) grains. Crop & Pasture Science, 68, 713–725.

Xiang, D., Yang, S., Li, X., Qian, Y., 2015. Life cycle assessment of energy consumption and GHG emissions of olefins production from alternative resources in China, Energy Conversion and Management, 90, 12-20

Xuan, D., Zhan, B., Sun Poon, C., 2018. A maturity approach to estimate compressive strength development of CO2-cured concrete blocks. Cement and Concrete Composites 85 (2018) 153-160.

Watson, A., Ghosh, S., Williams, M.J. *et al.* Speed breeding is a powerful tool to accelerate crop research and breeding. *Nature Plants* **4,** 23–29 (2018).

Yadav, S., Mehra, A. 2017. Experimental study of dissolution of minerals and CO2 sequestration in steel slag, Waste Management 64, 348–357

Yufang, T., Yongde, Z., Tao, H., Qiang, Z., Yongzhen, P. 2016. Preparation of lignin sulfonate-based mesoporous materials for adsorbing malachite green from aqueous solution, J. Environ. Chem. Eng. 4 (3) 2900–2910.